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AN INVESTIGATION ON PHOTOPHORESIS

A THESIS

Presented to

The Faculty of the Graduate Division

by

Edward Yun-ho Keng

In Partial Fulfillment

of the Requirements for the Degree

Master of Science in Chemical Engineering


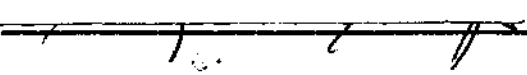
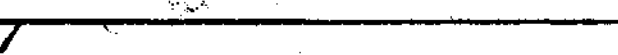
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AN INVESTIGATION ON PHOTOPHORESIS

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## TABLE OF CONTENTS

	Page
ACKNOWLEDGMENTS. . . . .	ii
LIST OF TABLES . . . . .	v
LIST OF FIGURES. . . . .	vii
SUMMARY. . . . .	xi
 Chapter	
I. INTRODUCTION . . . . .	1
Types of Photophoresis . . . . .	2
Historical Review. . . . .	4
II. THEORIES . . . . .	6
Radiometer Effect. . . . .	6
Electric and Magnetic "Ion" Effect . . . . .	13
Other Observations . . . . .	15
III. EXPERIMENTAL WORK. . . . .	17
Equipment. . . . .	17
Aerosol Chamber. . . . .	17
Optical System . . . . .	18
Shutter and Timer. . . . .	19
Vertical Displacement Measurement. . . . .	20
Electric Field Equipment . . . . .	20
Operation. . . . .	21
Calculation of Particle Size . . . . .	21
Calculation of the Photophoretic Force . . . . .	23
Mean Free Path of Gas Molecules and Slip Factor Evaluation. . . . .	23
IV. OBSERVATIONS AND RESULTS . . . . .	27
Convergent and Divergent Beams . . . . .	27
Projection Lamp as Source. . . . .	27
Focused Sunlight . . . . .	34

## TABLE OF CONTENTS (Continued)

Chapter	Page
Parallel Beams . . . . .	36
Stable Photophoresis . . . . .	36
Unstable Photophoresis . . . . .	37
Rising and Falling Velocities. . . . .	38
Electric and Magnetic Field Effects. . . . .	39
Wavelength Influence . . . . .	44
Analysis of Errors . . . . .	46
Method Errors. . . . .	46
Experimental Errors. . . . .	47
V. DISCUSSION OF RESULTS. . . . .	49
Comparison of Positive and Negative Photophoresis. . . . .	49
Dependence on Gas Medium . . . . .	50
Dependence on Light Intensity. . . . .	50
Effect of Particle Differences . . . . .	53
VI. RELEVANCE TO METEOROLOGICAL PHENOMENA. . . . .	69
Substances at High Altitudes . . . . .	69
Estimation of Atmospheric Particles Photophoresis. . . . .	71
VII. FINAL REMARKS. . . . .	79
Straight Path in Light's Direction . . . . .	79
Changing Directions. . . . .	79
Initiation of Photophoresis. . . . .	80
Boundary Phenomena . . . . .	81
Influence of Electric and Magnetic Fields. . . . .	81
Positive and Negative Photophoretic Forces . . . . .	81
Dependence on Particle Material and Gas. . . . .	82
VIII. CONCLUSIONS. . . . .	83
IX. RECOMMENDATIONS. . . . .	85
APPENDIX . . . . .	88
BIBLIOGRAPHY . . . . .	118

# LIST OF TABLES

Table	Page
1. Accommodation Coefficient of Platinum in Several Gases. . . . .	10
2. Experimental and Calculated Photophoretic Force Values . . . . .	12
3. Mean Free Path of Air and Helium Molecules . .	26
4. Comparison of the Properties of Air and Helium	40
5. Filters Used in Filter Test and Their Conditions. . . . .	45
6. Photophoretic Forces of Selenium Particles in Argon, Nitrogen, and Hydrogen Measured by Parankeiwicz . . . . .	53
7. Exponent on Particle Radius Term as a Function of Gas Pressure. . . . .	57
8. A Comparison of Photophoretic Force for Different Materials at a Pressure of 30 Torr. . .	62
9. Photophoretic Force as a Function of Gas Medium Pressures . . . . .	66
10. Estimated Values of Negative Photophoretic Forces in Stratosphere . . . . .	73
11. Estimated Values of Positive Photophoretic Forces in Stratosphere . . . . .	74
12. Estimated Rising and Settling Velocities of the Light-Negative Particles in the Stratosphere . . . . .	75
13. Estimated Falling Velocities of the Light-Positive Particles in the Stratosphere . . . .	76
14. Experimental Data of Positive Photophoretic Force for Sodium Chloride Particles. . . . .	89

## LIST OF TABLES (Continued)

Table	Page
15. Experimental Data of Negative Photophoretic Force for Sodium Chloride Particles . . . . .	90
16. Experimental Data of Positive Photophoretic Force for Silicon Carbide Particles . . . . .	92
17. Experimental Data of Negative Photophoretic Force for Silicon Carbide Particles . . . . .	93
18. Experimental Data of Negative Photophoretic Force for Carbon Particles in Air . . . . .	94
19. Experimental Data of Negative Photophoretic Force for Carbon Particles in Helium. . . . .	96
20. Experimental Data of Positive Photophoretic Force for Zinc Particles. . . . .	98



## LIST OF FIGURES

Figure	Page
1. Relations Between the Velocity of Particles and the Field Strength in Electrophotophoresis and Magnetophotophoresis. . . . .	3
2. A Force Analysis for a Rotating Particle in a Circular Orbit Motion . . . . .	29
3. A Force Analysis on a Particle Showing Vertical Ellipse . . . . .	30
4. Boundary Phenomenon of a Convergent Beam of Light . . . . .	32
5. Circular Orbit in Divergent Beam. . . . .	33
6. Force Analysis for Particle at the Edge of a Convergent Beam When the Light is from the Top . . . . .	36
7. Photophoresis and Electric Field. . . . .	41
8. Photophoresis and Magnetic Field. . . . .	42
9. Iron Particle Crossing the Beam in Magnetic Field . . . . .	44
10. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 1.5 Torr in Helium and Air. . . . .	51
11. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 10 Torr in Helium and Air . . . . .	52
12. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 30 Torr in Helium and Air . . . . .	52
13. Comparison of Positive Photophoretic Force for Gas Carbon Particles at a Pressure of 25 Torr Taken at Different Light Intensities. . . . .	54
14. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 10 Torr . . . . .	56

## LIST OF FIGURES (Continued)

Figure		Page
15.	Exponent on Particle Radius Term versus Gas Pressure as Shown in Table 7. . . . .	60
16.	Positive Photophoretic Force versus Particle Radius for Sodium Chloride, Silicon Carbide, and Zinc Particles at a Pressure of 30 Torr .	63
17.	Negative Photophoretic Force versus Particle Radius for Carbon, Sodium Chloride, and Silicon Carbide Particles at a Pressure of 30 Torr	63
18.	Negative Photophoretic Force versus Pressure for 0.38 Micron Radius Sodium Chloride Particle. . . . .	67
19.	Positive Photophoretic Force versus Pressure for 0.72 Micron Radius Silicon Carbide Particle. . . . .	67
20.	Negative Photophoretic Force versus Pressure for 0.5 Micron Radius Carbon Particle . . . .	68
21.	Negative Photophoretic Force versus Pressure for 0.9 Micron Radius Sodium Chloride Particle	68
22.	Falling Velocities for Particles with a Density of 2.0 gm/cm <sup>3</sup> versus Altitude. . . . .	78
23.	Negative Photophoretic Force versus Particle Radius for Sodium Chloride Particles at Pressures of 2 and 10 Torr. . . . .	99
24.	Positive and Negative Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 15 Torr. . . . .	100
25.	Positive Photophoretic Force versus Radius for Sodium Chloride Particles at a Pressure of 20 Torr. . . . .	101
26.	Positive and Negative Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 30 Torr. . . . .	102
27.	Positive and Negative Photophoretic Force versus Particle Radius for Sodium Chloride	

## LIST OF FIGURES (Continued)

Figure		Page
28.	Positive Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 63 Torr. . . . .	104
29.	Positive Photophoretic Force versus Particle Radius for Silicon Carbide Particles at a Pressure of 2 Torr . . . . .	105
30.	Positive and Negative Photophoretic Forces versus Particle Radius for Silicon Carbide at a Pressure of 5 Torr . . . . .	106
31.	Positive and Negative Photophoretic Forces versus Particle Radius for Silicon Carbide Particles at a Pressure of 10 Torr . . . . .	107
32.	Positive and Negative Photophoretic Force versus Particle Radius for Silicon Carbide Particles at a Pressure of 30 Torr . . . . .	108
33.	Positive and Negative Photophoretic Forces versus Particle Radius for Silicon Carbide Particles at a Pressure of 50 Torr . . . . .	109
34.	Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 0.6 Torr . . . . .	110
35.	Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 1.5 Torr in Air and Helium . . . . .	111
36.	Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 10 Torr in Air and Helium . . . . .	112
37.	Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 30 Torr in Air and Helium . . . . .	113
38.	Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 4 Torr . . . . .	114

## LIST OF FIGURES (Continued)

Figure	Page
39. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 10 Torr . . . . .	115
40. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 20 Torr . . . . .	116
41. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 30 Torr . . . . .	117

## SUMMARY

Photophoresis, a phenomenon involving the motion of particulate matter in a light beam, has been known and studied for about fifty years. Many aspects of this phenomenon have been found and discussed, but there is as yet no satisfactory theory that accounts for all the observed effects.

In this study of photophoresis many kinds of particles and a variety of conditions were investigated. Two possible explanations for this phenomenon--the radiometer force and "electric and magnetic charge" effects--were examined in particular. It was concluded that the radiometer force is very possibly the major cause of photophoresis, but that there must be some other interaction between small particles and a beam of light. Phenomena, such as the "funnel" effect, which had never before been described, were also uncovered in this study.

Both the positive and negative photophoretic forces exhibited by particles of several materials were measured and compared. Positive photophoresis is considered to be that displayed by particles that move away from the light and negative photophoresis by those that move toward the light. Overall, most of the positive photophoretic force values are of the same order of magnitude as the negative

ones. No differences in behavior were observed between photophoretic phenomena in air and in helium. The photophoretic force was measured in both air and helium and no significant difference was found here. Electric and magnetic fields were employed in this study in order to reveal the nature of photophoresis. Results suggest that the behavior of particles in this situation arises from the combination of two or more effects. There is no evidence that light-positive particles and light-negative particles carry different charges. A "funnel" effect was observed in a focused beam of sunlight directed downward. The particles at the boundary of the converging portion of the beam moved along the boundary without falling from the beam. The phenomenon is very similar to the motion of particles in a liquid passing through a funnel.

Predictions of particle behavior in the upper atmosphere were made by relating measured photophoretic force values to gas pressure, light intensity, and particle properties at several levels. Photophoresis is shown to be capable of causing some very small particles to rise and some to fall faster under gravity than they normally would. It is speculated that this may have significant consequences in regard to meteorological processes and fallout behavior.

## CHAPTER I

### INTRODUCTION

The phenomenon of motion exhibited by small, gas-suspended particles in an intense beam of light under reduced pressure is called photophoresis. When closely observed, particles may be seen both to rotate and to undergo translational motion as long as the light is on. The movement may be in a straight line, a circle, a helix, or an irregular curve, and the particles may move either away from or toward the light source. Particles that move away are called light-positive, and those that move toward the light are called light-negative. These two different phenomena, light positive and negative, are observed simultaneously in most cases when several particles of a single substance are visible. Sometimes both actions occur at different times with a single particle, i.e., a light-positive particle may change to a light-negative one or vice versa. Straight-line motion prevails in a parallel beam of light, while circular, helical, or irregular motions are characteristic of divergent or convergent beams. In the converging section of a focused beam of sunlight, particles have been observed to "bounce" off the beam's outer boundary as though it were actually a solid wall.

This indicates the special nature of the light-dark interface.

This investigation was first undertaken by examining the photophoretic behavior of particles in both parallel beams and cone-shaped beams. Particles were observed under pressures from 0.5 torr to atmosphere pressure. Photophoretic forces were measured for several substances by suspending the particles in a known-intensity, more-or-less parallel beam. Both positive and negative photophoretic forces were measured and compared. Air and helium were used as the gaseous media; unless otherwise specified, air alone served as the gaseous medium.

The object of this study was to predict particle behavior in the upper atmosphere by relating photophoretic force to gas pressure, light intensity, and particle properties.

#### Types of Photophoresis

Photophoresis is reported to occur both in gases and liquids<sup>(3)</sup>, but only gas-borne particle photophoresis will be considered here in any detail. There are several types of gas-borne photophoresis. They may first be divided simply into two groups. One is called pure photophoresis and the other, field photophoresis. Pure photophoresis is exhibited in the presence of the gravitational field which apparently has no influence; it is produced simply by light



only. The pressure of the gas medium, the intensity and wave length of the light beam, and the nature of the particles are the main factors influencing this type. Field photophoresis involves either an electric field (electrophotophoresis), a magnetic field (magnetophotophoresis), or both (electro-magnetophotophoresis) and pure photophoresis. (14)(25)(46)(47) More than two fields present simultaneously create what is called complex photophoresis. Field photophoretic forces are proportional to the strength of the field for low strengths, but they will reach a saturated value at higher fields. (15)(25)(46) In other words, once a saturation value is attained, the force no longer depends on the field strength. Typical relationships between the velocity of particle motion and field strength for electrophotophoresis and magnetophotophoresis (15)(46)(47)(48) are plotted in Figure 1.

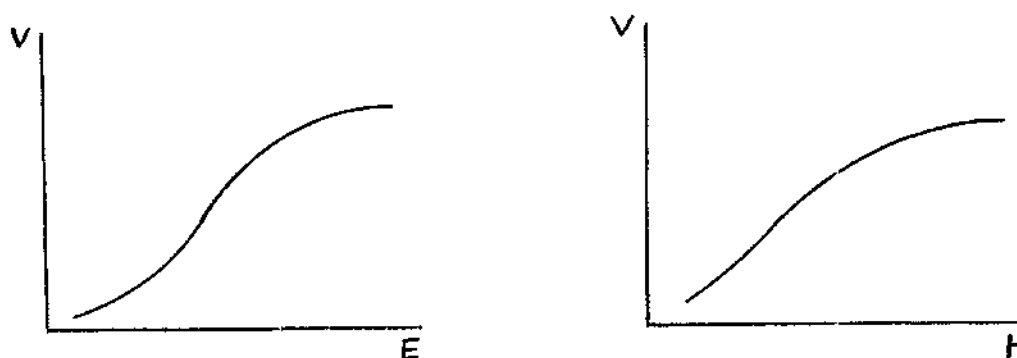


Figure 1. Relations between the Velocity of Particles and the Field Strength in Electrophotophoresis and Magnetophotophoresis. (V is particle velocity. E and H are the strength of the electric and magnetic fields, respectively.)

Pure photophoresis and field photophoresis are compared further by the following statements.

<u>Pure Photophoresis</u>	<u>Field Photophoresis</u>
1. The motion of particles depends on the direction of the light beam.	1. The motion of particles depends on the direction of the field, but not on the light beam direction.
2. The photophoretic force is strongly dependent on the intensity of the light beam.	2. The photophoretic force is strongly dependent on the intensity of the light beam, but, also, the strength of the field in weak fields (saturation occurs in stronger fields).
3. The rotation of particles is about the direction of light as an axis. (25)(47)	3. The rotation of particles is about the direction of the field as the axis. (25)
4. The photophoretic force varies with variation in gas pressure.	4. A similar dependence on pressure is also observed in field photophoresis. (46)
5. Photophoresis disappears if the light is cut off.	5. Field photophoresis disappears when either the light or the field is cut off.

#### Historical Review

Photophoresis was discovered by F. Ehrenhaft<sup>(11)</sup> about 50 years ago. The phenomenon has since been observed by many investigators, but a completely satisfactory theory for it has not yet been found. Photophoresis was attributed to radiometer effects from the first. In 1926, Hettner<sup>(28)</sup> derived a theoretical equation to calculate the photophoretic force acting on the particle by assuming that

photophoresis was a radiometer effect. The agreement between some experimental data and theoretical values gives strong support to this assumption. About the same time, Barkas<sup>(3)(4)</sup> found that the photophoresis phenomenon also existed in liquids, and that X-rays could cause photophoresis.<sup>(3)</sup> Measured forces for silver particles in a liquid<sup>(4)</sup> were of the same order of magnitude as the values measured by Ehrenhaft<sup>(12)</sup> in nitrogen gas. Since a radiometer force cannot be developed in a liquid due to the short mean free paths of liquid molecules, the radiometer effect can hardly account for this observation. Some other phenomena observed in air under low pressures also cannot be explained as due to a radiometer effect. An example is field photophoresis, which depends on the intensity of the light but not its direction. In 1942, Ehrenhaft advanced the idea that photophoresis was due to "electric and magnetic ions."<sup>(19)</sup> This explanation may account for many of the observed photophoretic phenomena if, as Ehrenhaft said, the light induces electric and magnetic charges (poles) upon the particles when they are illuminated by concentrated light of preponderantly the shorter wave lengths, but it is not clear how the photophoretic force develops. Even though the radiometer effect is not satisfactory to explain all the phenomena associated with photophoresis, most investigators still favor it.<sup>(1)(8)(26)(28)(37)(45)(46)(47)(57)(59)(61)</sup>

## CHAPTER II

### THEORIES

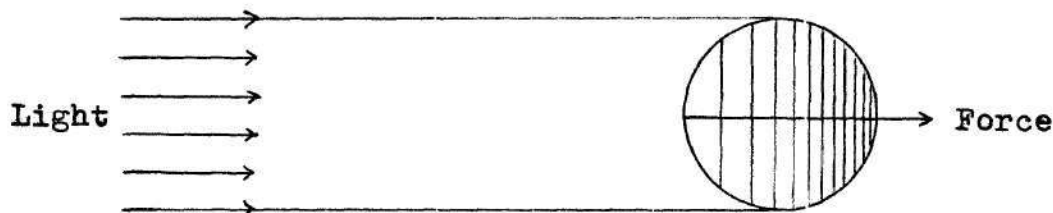
There is as yet no single theory of photophoresis that accounts for all the observed effects. Two, however, that are much more successful than any of the others will be considered here. The first attributes photophoresis to a radiometer effect and the second to an electric and magnetic "ion" effect.

#### Radiometer Effect

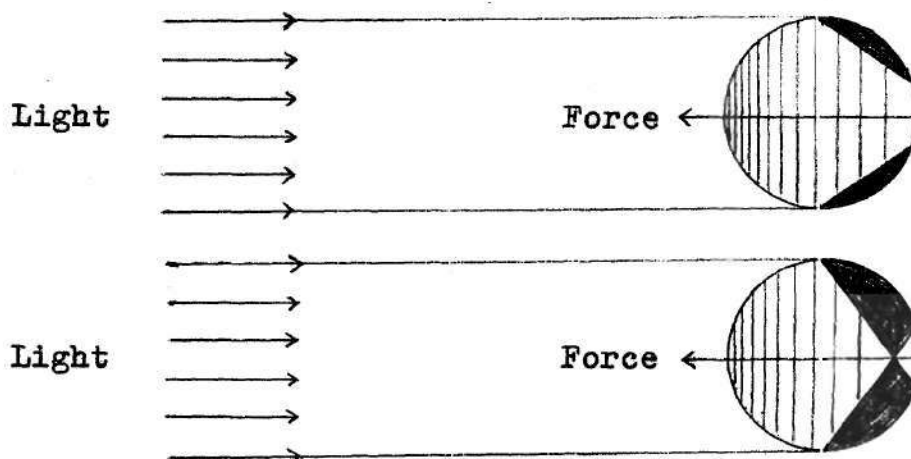
Since small particles irradiated by a strong beam of light show both light-positive and light-negative motion, an interpretation by light pressure cannot be accepted. Radiometer effect then becomes the favored theory with which to interpret photophoresis even though some of the observed phenomena cannot be explained by it. The development of a radiometer force requires that the surface of a particle be unevenly heated. The temperature difference is produced by the light radiation and its magnitude depends on the size, shape, and nature of the particles. Gas molecules arriving at the hottest portion of the particle surface have velocities appropriate to their original temperature. As result of the collision, energy is absorbed and their rebound velocities are increased. The uneven incre-

ment in the velocities from the hotter and cooler surface parts will cause a reaction force. Several investigators (15)(47)(52) have postulated how the surface of a spherical particle might be unevenly heated. Their main arguments may be summarized as follows:

1. Light-positive motion: The front, or exposed, side of the particle is heated directly by the light, and radiation energy is absorbed and emitted by the front side only.



2. Light-negative motion: The back, or unexposed, side of the particle is heated more effectively by the concentration of light through the translucent body of the particle.



The radiometer force depends also on the accommodation coefficient  $\gamma$  defined as<sup>(35)(47)</sup>:

$$\gamma = \frac{T_b - T_a}{T_s - T_a} \quad (1)$$

where  $T_a$  and  $T_b$  are the temperatures of the gas molecule before and after hitting the wall, respectively.  $T_s$  is the temperature of the surface where the gas molecules are hitting. The accommodation coefficient  $\gamma$  depends on the nature of the surface of the body as well as the gas molecules themselves. A body having a changing accommodation coefficient across its surface may show a radiometer force even with a uniform temperature.

The radiometer force acting on a spherical particle will, according to theory, reach a maximum value at the pressure  $P_{\max}$ , when the mean free path of the gas molecules is of the same order of magnitude as the sphere radius. For particle radii smaller than the mean free path of the gas molecules, the radiometer force is proportional to the gas pressure, and for particle radii larger than the mean free path of the gas molecules, the radiometer force is inversely proportional to the pressure.<sup>(28)(47)(61)</sup>

Hettner<sup>(28)</sup> derived an equation based on the radiometer effect for the pressure  $P_{\max}$  at which the maximum force exists. The equation is

$$P_{\max} = \frac{3\mu}{a} \sqrt{\frac{RT}{\gamma M}} \quad \text{or} \quad P_{\max} = 2.34 \frac{P\lambda}{a\sqrt{\gamma}} \quad (2)$$

where  $\gamma$  is the accommodation coefficient,  $\mu$  is the gas viscosity,  $P$  and  $T$  are the pressure and temperature of the gas,  $a$  is the particle radius, and  $\lambda$  is the mean free path of the gas molecules.

The values of  $P\lambda$  are quite constant. According to an equation given later, equation (21), the value of  $P\lambda$  for air may be given as

$$P\lambda = 5.09 \times 10^{-3} \quad (3)$$

then equation (2) for air becomes

$$P_{\max} = \frac{11.91}{a\sqrt{\gamma}} \times 10^{-3} \quad (4)$$

where  $P$  and  $P_{\max}$  are in torr,  $\lambda$  and  $a$  are in cm, and  $\gamma$  is dimensionless. The values of  $\gamma$  for different solids and different gases are not the same, and adequate data for could not be found in the literature. Some measurements by Knudsen<sup>(35)</sup> indicating the dependence of the accommodation coefficient on the gas and surface conditions are listed in Table 1.

Hettner<sup>(28)</sup> derived the equation for the photophoretic force in relation to the accommodation coefficient

$$F_p = 2.86 \frac{P \Delta T}{T} \cdot f \quad (5)$$

where  $f$  is given by  $\lambda^2 / 1 + (5.5 \lambda^2 / \gamma a^2)$  and  $\Delta T$  is the temperature difference between opposed points on the

Table 1. Accommodation Coefficient of Platinum  
in Several Gases

Platinum Condition	Accommodation Coefficient in the Gases		
	Hydrogen	Oxygen	Carbon Dioxide
Bright	0.358	0.835	0.868
Thin Platinized	0.556	0.927	0.945
Thick Platinized	0.712	0.956	0.975

illuminated and the nonilluminated parts of the particle.

Hettner<sup>(28)</sup> has also derived an equation for pressures higher than  $P_{\max}$ , describing the radiometer force for spheres by

$$F_R = \frac{3 \pi R \mu^2 \Delta T}{2 MP} \quad (6)$$

He has also given an equation for pressures lower than  $P_{\max}$  based on an equation originally derived by Rubinowitz<sup>(51)</sup>, describing the radiometer force for spheres by

$$F_R = \frac{\pi \gamma P a^2 \Delta T}{6 T} \quad (7)$$

For the transition region ( $\lambda$  comparable to  $a$ ), he gave

$$F_R = \frac{\pi \mu \left( \frac{R}{4 MT} \right)^{1/2} a \Delta T}{\frac{P}{P_{\max}} + \frac{P_{\max}}{P}} \quad (8)$$

When the pressure and temperature of the gaseous medium are constant, equation (6) can be written as:

$$F_R = C \Delta T, \quad (9)$$



where

$$C = \frac{3 \pi R \mu^2}{2 MP} = \text{constant} \quad (10)$$

So, for pressures higher than  $P_{\max}$ , the radiometer force acting on a spherical particle is proportional to  $\Delta T$ . But for pressures lower than  $P_{\max}$ , the radiometer force acting on a spherical particle is proportional to  $a^2 \Delta T$  according to equation (7).

Weber<sup>(60)</sup> derived an equation describing  $\Delta T$  in terms of particle and medium thermal conductivities when the particle radius is larger than the mean free path of the gas molecules, i.e., under pressures higher than  $P_{\max}$ . The equation is

$$\Delta T = \frac{2 I a}{X_p + 2X_g} \quad (11)$$

where  $I$  is the intensity of the light beam and  $X_p$  and  $X_g$  are the thermal conductivities of the particles and gas medium, respectively. In most cases,  $X_p$  is much larger than  $X_g$ , so that  $\Delta T$  depends strongly on  $X_p$ . According to the experimental results, the photophoretic force does not depend strongly on the thermal conductivity. In other words,  $\Delta T$  cannot be expressed only in terms of light intensity, particle radius, and thermal conductivities. It should include radiation energy absorption and light reflection and refraction.

Substituting equation (11) into equation (5) and calculating the photophoretic force for a sodium chloride particle at 50 torr, the results are as shown in Table 2.

Table 2. Experimental and Calculated Photophoretic Force Values

Particle Radius (micron)	Experimental Photophoretic Force Values (dyne)	Photophoretic Force Values Calculated by Equations (5) & (11) (dyne)
0.5	$1.1 \times 10^{-8}$	$3.0 \times 10^{-10}$
1.0	$7.0 \times 10^{-8}$	$2.1 \times 10^{-9}$
2.0	$4.3 \times 10^{-7}$	$1.2 \times 10^{-8}$

The big difference between the smoothed and calculated values shown above may be due to the evaluation of  $\Delta T$ , the expression which was derived for a higher pressure region. A pressure of 50 torr and particle radii as shown above locates the system in the transition region, i.e., in a region where the mean free path of the gas molecules is comparable to particle radius  $a$ . It is surprising in fact that the exponent on the particle radius term (slope from a log-log plot of photophoretic force versus particle radius) from calculated values agrees as well as it does with values from experimental results. The two are 2.75 and 2.69, respectively. Equation (6) is for pressures higher than  $P_{\max}$ . Due to poor experimental data in this region, no good comparison can be made at high pressures.

Due to the lack of data for accommodation coefficients and the fact that no practical method is available for calculating  $\Delta T$  in the low pressure region for different particles and gases, there is no way to compare equations (5), (6), (7), and (8) with experimental measurements. It may be noted, however, that according to equation (5),  $f$  does not strongly depend on the gaseous medium in the case of platinum where values of the accommodation coefficient and mean free paths of gas molecules are available. The thermal conductivities of the particle are much larger than those for gases. So  $\Delta T$  depends strongly on the particles themselves but weakly on the gaseous medium. This may explain why the same magnitude photophoretic forces were measured in different gas mediums by Parankiewicz<sup>(44)</sup> and in this study more information on  $\gamma$  and  $\Delta T$  are needed.

#### Electric and Magnetic "Ion" Effect

Some phenomena associated with photophoresis cannot be explained sufficiently as a radiometer effect. For example, the same kinds of particles under the same gas pressure show both light-positive and light-negative photophoresis in the same beam of light and the positive and negative forces are found to be of almost the same magnitude. Barkas<sup>(3)(4)</sup> also found both light-positive and light-negative photophoresis in liquids where the

radiometer force could not exist due to the short mean free path of the liquid molecules. Field photophoresis depends on the intensity of the light beam, but not its direction. Since so many phenomena in photophoresis are not explained by the radiometer effect, some investigators have tried to interpret it through the nature of light. Up to the present, however, no satisfactory light-oriented theory of photophoresis has been obtained.

Ehrenhaft<sup>(15)(22)</sup> reportedly found that a charge apparently smaller than that of a single electron exists on small spheres. The ratio of this charge to that of the electron was measured later by Tausin<sup>(56)</sup> to be 0.2. In 1941, Ehrenhaft<sup>(19)</sup> interpreted photophoresis by what he called electric and magnetic "ions." He stated that intense light of relatively shorter wave lengths could induce electric and magnetic "charges" on the particle. He maintained that not only field photophoresis but also pure photophoresis was really a manifestation of electromagnetic phenomena, and he believed that there existed a stationary electrostatic field in the direction of propagation of a beam of light.<sup>(16)(17)</sup>

This interpretation of photophoresis, if accepted in its entirety, can explain most of the phenomena which the radiometer effect cannot--such as photophoresis in liquid and field photophoresis. Positive- and negative-photophoretic forces are thus due to the different charges

induced by light on them, which accounts for their having forces of the same order of magnitude. The light can induce charges on particles<sup>(19)(32)</sup> and also can change them. This explains why some particles change from light positive to light negative and vice versa. Since this interpretation requires no heating effects, photophoretic phenomena in liquid caused by X-rays<sup>(3)</sup> are a possibility.

Regular and irregular curved motions are observed in convergent beams. This raises many questions which are not easily explained by electric and magnetic ions. Pure photophoretic forces are developed between the electric and magnetic ions and the light itself if the interpretation by electric and magnetic ions is accepted. A further study on the nature of convergent-beam photophoresis is necessary before phenomena observed here are resolved. Some of these phenomena are discussed in a later section. Because photophoresis is found with considerably longer wavelength light than ordinarily is considered to produce ionization, its effect can hardly be interpreted by charge induction on particles such as metals, crystals, and so on.

#### Other Observations

Various phenomena have been observed by many investigators and quite a few of the results are contradictory. Barkas<sup>(3)(4)</sup> found photophoresis with particles in a liquid and even produced it by X-rays. Quang Te-Tchao<sup>(42)(43)</sup>

attached light-negative particles between films and fixed the assembly to a freely rotating scale. The whole system was evacuated to  $5 \times 10^{-6}$  torr and then illuminated by a beam of light. The mica films with light-negative particles moved toward the light. This was interpreted as indicating that photophoresis was not a heating effect. Tauzin<sup>(58)</sup> and Robatschek<sup>(47)</sup> questioned the ascertainment, however.

Ehrenhaft interpreted photophoresis by electric and magnetic ions<sup>(19)</sup> as described above. He reported the existence of single magnetic poles or unipolar magnetic charges<sup>(18)(20)(23)(24)</sup> having values of  $10^{-8}$  to  $10^{-12}$  emu for single nickel and iron particles.<sup>(19)(24)</sup> He believed that the light induced unipolar magnetic charges on particles and these, in turn, produced photophoretic phenomena.<sup>(17)(19)</sup> A test for the existence of single magnetic poles was made by Benedikt and Leng.<sup>(5)</sup> The test was performed with a colloidal solution of  $\text{Fe}_3\text{O}_4$  and with fine iron and nickel particles. They concluded that no motion such as would be expected from single magnetic poles was in evidence and that an analysis of the results showed no evidence for the existence of single magnetic poles exceeding  $1.5 \times 10^{-12}$  emu.

## CHAPTER III

### EXPERIMENTAL WORK

#### Equipment

The overall apparatus was shown in a previous paper<sup>(49)</sup>; details of its various parts were also shown there by detailed drawings. Its basic components were (1) an aerosol chamber in which gas-suspended particles were exposed to the beam from one of several lamps, (2) a solenoid-operated shutter activated and timed by a thyatron device, and (3) a slide wire variable resistor adapted to record the vertical displacement of the particles. These components are discussed in more detail below.

#### Aerosol Chamber

This chamber, which is made of 1/8 inch thick steel, is approximately 1 foot long and has a square cross section that is 3 inches on a side. At the top and bottom of the chamber are removable steel plates. One of them is fitted with a compound lens to allow entrance of a light beam. There are three brass pins on the other plate that fit tightly into recesses in the base on which the chamber rests and to which is mounted the light source. These pins ensured replacement of the chamber in the same position relative to the light beam each time it was removed. The

chamber also has an 8-3/4 by 1/4-inch observation slit along one side covered by a Plexiglass window and marked with a measuring scale. The inside of the chamber is painted black to minimize scattered light. There are two 5/8-inch holes in two side walls through which electrodes may be mounted when needed. The chamber could be mounted vertically, with either end uppermost so that both negative and positive photophoresis could be opposed by gravity.

#### Optical System

Three light sources were used in this investigation. One was a 100-watt zirconium arc lamp, manufactured by Sylvania Electric Products, Inc., New York, New York. This lamp required 15.4 volts direct current at 6.25 amperes and a momentary starting potential of 2000 volts obtained from a power supply, model G-157, of the Gaertner Scientific Corporation, Chicago, Illinois. Another light source was an 80-watt high pressure mercury arc lamp, manufactured by Pek Labs, 4024 Transport Street, Palo Alto, California. The same power supply was used with both light sources. The third light source was an incandescent movie projection lamp. Two convex lenses forming a converging-diverging beam were used with them. This system was employed only for general observation; the other two were used for the detailed analysis because more nearly uniform light beams could be formed with them. The sun itself was used in a few experiments.



In order to produce an intense parallel beam an auxiliary optical system is required. Several lens combinations were tested, with the final system being built around two matched achromatic lenses, each having a 1-1/2-inch diameter and a 3/4-inch focal length. The lenses were mounted in threaded steel casings. One of the lens units was fixed onto the aerosol chamber, as noted above, and the other was mounted on a frame provided with a screw-type, fine adjustment. The frame was also movable for coarse adjustment.

For measuring the absolute intensity of the light beam an Eppley Laboratories Inc., Newport, Rhode Island, 12-junction, iron-constantan thermopile was used in conjunction with a model 8686 potentiometer of the Leeds and Northrup Company, Philadelphia, Pennsylvania.

#### Shutter and Timer

Interruption of the light beam to obtain up-and-down particle movement was accomplished with two solenoids and a metal blade assembly. Activation of one solenoid caused the beam to be obstructed, and the other removed the obstruction. In order to time these intervals accurately, an adaption of a thyatron device<sup>(55)</sup> was employed. Two units were coupled together, each operating one of the solenoids, such that activation of one and deactivation of the other occurred simultaneously. The activation times were established by rheostats in the timer. The shutter could

be opened and closed continuously at constant time intervals with an accuracy of  $\pm 0.05$  second without the necessity of individually timing each cycle.

#### Vertical Displacement Measurement

To measure the distance of rise and fall of the particles showing the photophoretic effect, a slide-wire resistor was fitted with a pointer and mounted so that a particle moving in the chamber could be followed with the pointer. By impressing a constant voltage across the resistor, changes in particle height in the chamber are thus translated into changes in electromotive force, the latter being recorded on a model S-72150 recorder of the E. I. Sargent and Company, Chicago, Illinois. In this manner, distances of rise and fall were measured as the light beam was interrupted and reinstated by merely setting the pointer at the upper and then the lower positions assumed by a particle.

#### Electric Field Equipment

In order to check the nature of light-positive and light-negative particles, equipment was installed with which to introduce an electric field. Two copper plates 10 inches long and 1 inch wide were used as electrodes. The distance between the two electrodes could be adjusted from less than 1 inch to 2 inches. A direct current power supply was employed to produce an impressed potential of up to 20,000 volts across the electrodes.

### Operation

The apparatus was designed so that only one person was needed to operate it. An experiment was initiated by putting a small quantity of dry powder in the aerosol chamber, sealing it, evacuating, and then agitating to make some of the particles become airborne. A single particle caught in the light beam and showing photophoresis was next selected for observation. The beam shutter was started and times were adjusted so that the particle rose and fell in as nearly as possible the same position in the chamber. The particles' terminal positions, finally, were located and recorded with the pointer. Shutter-open and shutter-closed time intervals were also recorded. The distance of rise and fall of the particles was also read directly from the scale on the viewing window when time permitted.

### Calculation of Particle Size

A particle moving in a viscous medium under the influence of a constant force reaches a constant linear velocity,  $V$ , that is determined by the force. This relationship may be expressed by

$$V = FZ \quad (12)$$

where  $Z$ , the mobility<sup>(38)</sup> of the particle, is characteristic of particle size and shape and of the properties of the fluid medium. For a sphere in a homogeneous medium, the

mobility is given by

$$Z = \frac{1}{6 \pi \mu a} \quad (13)$$

a form of Stokes' law where  $\mu$  is the viscosity of the medium and  $a$  the particle radius. As the mean free path becomes comparable to the particle size, equation (13) must be modified by inclusion of a slip correction factor  $S$ . The modified mobility,  $Z_m$ , is then defined by

$$Z_m = SZ = \frac{S}{6 \pi \mu a} \quad (14)$$

The slip correction, a function of  $\frac{\lambda}{a}$ , which is the ratio of molecule mean free path to the particle radius, accounts for the tendency of a particle to slip between molecules of the gaseous medium. (9)(10)(40)

When the velocity in equation (12) is the settling velocity,  $V_s$ , the force becomes the gravitational force acting on the particle defined by

$$F_g = \frac{4}{3} \pi a^3 g \rho_p \quad (15)$$

where  $g$  is the gravitation constant, and  $\rho_p$  is the particle density. Substituting and solving for  $a$  gives

$$a = \sqrt{\frac{9 V_s \mu}{2 S g \rho_p}} \quad (16)$$

The velocity of fall under gravity was thus employed to establish particle radii.

#### Calculation of the Photophoretic Force

Having determined the particle radius, the photophoretic force,  $F_p$ , was evaluated by

$$F_p = F_d + F_g \quad (17)$$

where  $F_d$  is the drag force exerted by the fluid on the rising particle. Substituting equations (12), (14), and (15) into equation (17) gives

$$F_p = \frac{6 \pi \mu a V_r}{S} + \frac{4}{3} \pi a^3 g \rho_p \quad (18)$$

where  $V_r$  is the velocity of the rising particle.

#### Mean Free Path of Gas Molecules and Slip Factor Evaluation

In order to apply the equations developed above, the slip factor must be evaluated. The slip effect, as mentioned in the preceding sections, was investigated by Knudsen and Weber<sup>(36)</sup>, who derived an empirical relationship defining the slip factor as:

$$S = 1 + \frac{\lambda}{a} \left[ A + B e^{-\frac{Ca}{\lambda}} \right] \quad (19)$$

According to an analysis by Davies<sup>(10)</sup>, the best values of the empirical constants A, B, and C are  $A \approx 1.257$ ,  $B = 0.400$ , and  $C = 1.10$ . The mean free path,  $\lambda$ , is given by

$$\lambda = \frac{\mu}{0.499 \rho_g \bar{u}} \quad (20)$$

where  $\rho_g$  is the density of the gas medium and  $\bar{u}$  is the mean molecular speed. At low pressure, if ideal gas behavior is assumed,  $\lambda$  can be written in a more convenient form as

$$\lambda = \frac{\mu RT}{0.499 M P \bar{u}} \quad (21)$$

where R is the universal gas constant, T is the absolute temperature, M the molecular weight of the gas, and P is the absolute pressure.

According to the kinetic theory of gases, the mean molecular speed of gases,  $\bar{u}$ , is given by

$$\bar{u} = \sqrt{\frac{8 k T}{\pi m}} = \sqrt{\frac{8 R T}{\pi M}} \quad (22)$$

where k is the Boltzmann constant and m is the mass of a single gas molecule.

From the experimental results measured by Kestin and Leidenfrost,<sup>(33)</sup> the viscosities of gases may be assumed to be constant in the low pressure range at constant tempera-

ture. The values of mean free path calculated by equation (21) agree very well with those given by Orlicek<sup>(41)</sup> and by Havens, Koll, and LaGow.<sup>(27)</sup> The comparison values are listed in Table 3.

Table 3. Mean Free Paths in Air and in Helium

<u>P (Torr)</u>	<u>T (°K)</u>	<u><math>\lambda</math> (cm) by Eq. (21)</u>		<u><math>\lambda</math> (cm) by Havens, et al (N<sub>2</sub>)</u>	
760	290	$6.53 \times 10^{-6}$		$6.5 \times 10^{-6}$	
210	230	$1.87 \times 10^{-5}$		$1.9 \times 10^{-5}$	
42	210	$8.55 \times 10^{-5}$		$8.6 \times 10^{-5}$	
9.5	235	$4.23 \times 10^{-4}$		$4.2 \times 10^{-4}$	
2.4	260	$1.85 \times 10^{-3}$		$1.8 \times 10^{-3}$	
<u>P (Torr)</u>	<u>T (°K)</u>	<u><math>\lambda</math> (cm) by Eq. (21)</u>		<u><math>\lambda</math> (cm) by Orlicek</u>	
		<u>Air</u>	<u>He</u>	<u>Air</u>	<u>He</u>
1	293	$5.01 \times 10^{-3}$	$1.42 \times 10^{-2}$	$4.9 \times 10^{-3}$	$1.4 \times 10^{-2}$
0.5	293	$10.02 \times 10^{-3}$	$2.84 \times 10^{-2}$	$9.8 \times 10^{-3}$	$2.8 \times 10^{-2}$



## CHAPTER IV

### OBSERVATIONS AND RESULTS

#### Convergent and Divergent Beams

Many kinds of particle have been observed showing irregular path photophoresis in convergent and divergent light beams. Sunlight and a movie projection lamp were used for the light sources.

#### Projection Lamp as Source

When a projection lamp was used, the light beam was introduced into the chamber from the bottom and focused at a point near the center. A powerstat was used to vary and control the intensity of the light beam. Many kinds of particles were examined, and carbon particles were found to be best for observation. The air pressure in the chamber was varied from 0.5 torr to atmosphere pressure, but pressures of from 5 torr to 15 torr were found to be the best region to see the phenomena of photophoresis. At pressures higher than 15 torr air convection currents become noticeable. For high density or large size particles, a higher pressure may be used.

Most particles were observed to move toward the focus, so this is understood to mean that the photophoretic force was acting in the direction of the light beam.

While some particles travelled with curved paths, including a helix, horizontal circle, vertical ellipse, irregular closed curve, or irregular curve with no fixed path, other particles stayed at a fixed point at the edge of the beam and rotated. All these phenomena were due to photophoresis because they disappeared when the light was cut off. They were strongly dependent on the intensity of the light. Motions in opposite directions that occurred in the same region were observed. This proved that convection currents are not the driving force for the particles.

The phenomena described above may be interpreted as a radiometer effect if consideration is given to the irregular shape of the particles. If such a particle rotates about its own axis so that the force on its surface is changing direction, then it may transcribe a circular or elliptical orbit. At the position where the vertical component of the photophoretic force just equals the gravity force, a horizontal circular orbit may develop. Figure 2 is intended to give a clearer view of this mechanism. If  $F_g = F_v$ , then there is no vertical motion.

It has been stated that the particle rotates one revolution simultaneously with the completion of one helix. (25) This gives very strong support to the above interpretation.

When the intensity of the beam was changed, the path of a particle describing a vertical ellipse often

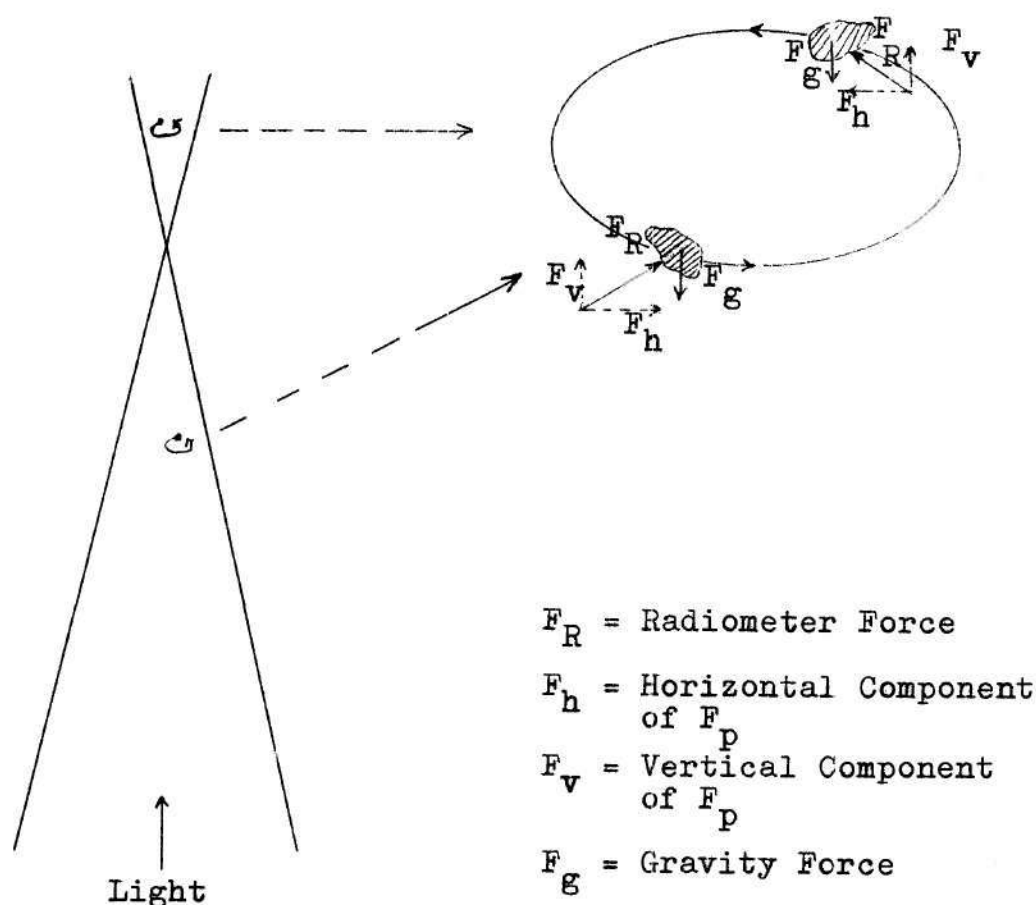


Figure 2. A Force Analysis for a Rotating Particle in a Circular Orbit Motion

shifted. A particle ordinarily went up inside the beam and down outside of it. The orbit circumference may be longer than 1 inch or shorter than 1/8 inch. When the path is short, the particle rises and descends along a steeper path. Varying the intensity of the light will cause a change of position, frequency, and length of the path. The motion will return to its original form if the intensity is changed back to its original intensity slowly. The path and the force analysis for this situation is shown in Figure 3.

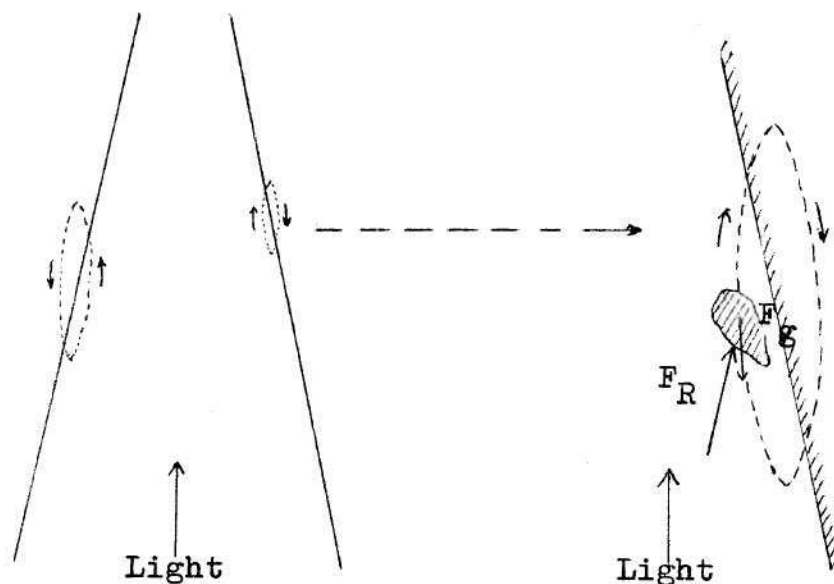


Figure 3. A Force Analysis on a Particle Showing Vertical Ellipse

$F_R$  = Radiometer Force

$F_g$  = Gravity Force

Air convection at the beam edge probably combines with the photophoretic (changing direction with rotation) and gravity forces to produce the vertical elliptical motion. A particle will rotate with its own axis at a fixed point in the edge of the beam if these forces compensate for one another and thus lead to an equilibrium situation. This case has been observed very often. Varying the intensity of the beam may destroy the equilibrium. The particle may rise or descend to obtain another equilibrium or start a vertical elliptical motion. Sometimes it may drop out of the beam.

Many different phenomena have been seen in a non-parallel beam that cannot be explained through electric and magnetic charges. Ehrenhaft<sup>(19)</sup> was quite confident

of the correctness of his interpretation of photophoresis by electric and magnetic ions in 1941 and 1942. He could explain then all the phenomena he had found in longitudinal photophoresis. But 10 years later, in 1951, he found that there was no way to explain phenomena such as described in this section. He concluded that it was impossible to explain the various observations by the knowledge at that time.<sup>(25)</sup>

Of the particles exposed to a convergent beam of light, some may move vertically or horizontally, some remain stationary but rotate about a central axis, and others rotate while moving in closed orbits. Several different kinds of motion happen simultaneously in the same region of the beam. This indicates that air convection currents are not important in those phenomena even though they may have a little influence.

Once two carbon particles were observed travelling horizontally close to the focus of a vertical divergent beam. Both of them moved in orbits which were like triangles with  $1/4$ -inch sides. The upper particle's orbit was larger than the lower one and both of them had a speed of about one half revolution per second, but with opposite directions. The distance between the two horizontal triangular paths was about  $1/4$  inch. The lower path sometimes turned to an irregular closed curve. This phenomenon was observed for about half an hour. In the same beam, a

particle was found reciprocating along a nearly straight path about 1 inch long. When the particle reached the upper end of the path, it turned back very fast, just as if it had been struck by an invisible bat. Then it slowed until it reached the lower end of its path. There it turned back and rose again. Each round trip took about 3 seconds. This motion was observed for about 20 minutes, and when the intensity of the light was lowered, the particle dropped from the beam.

In the same beam as used before--a convergent one aimed from the bottom upward--a special phenomenon was observed at the boundary. Carbon particles fell in the unlighted zone, as shown in Figure 4.

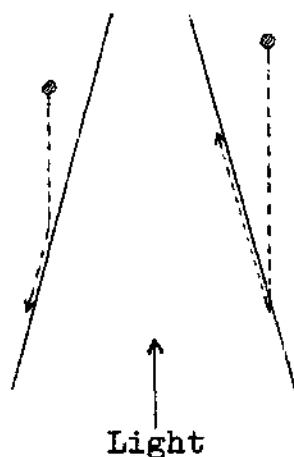


Figure 4. Boundary Phenomenon of a Convergent Beam of Light

They were visible by reflected light even when not in the beam itself. When they reached the boundary of the cone-shaped beam, some particles were pulled into the beam and thus pushed up by the light along the boundary. Some of them were stopped at the boundary and slid down along the boundary. This phenomenon is very similar to the funnel effect observed in sunlight, which is discussed in the next section.

Horizontal circular orbits were observed in divergent beams above the focus, as shown in Figure 5. Sometimes irregular closed curves were also observed. The

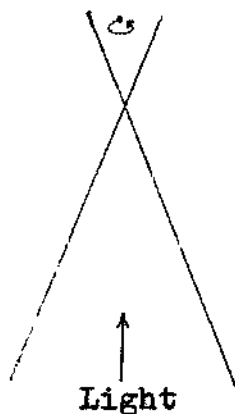


Figure 5. Circular Orbit in Divergent Beam

particles could be kept in the beam for hours. Varying the intensity would vary the height of the path. In general, the height of the path was proportional to the intensity. Once when the pressure was 5 torr, a carbon particle was observed moving along a 1/16-inch-diameter circular orbit. The speed was so fast that it looked like a white

ring. When the intensity of the light was varied, the height of the path changed while the diameter of the circular path stayed the same. There was a lower limit of light intensity. Below the limit, a particle would fall through the focus and move toward the bottom of the chamber by a helical path. When the intensity was switched higher soon after the particle passed the focus, the particle would rise, passing the focus and attaining the original path again. The force analysis is shown in Figure 2.

#### Focused Sunlight

Photophoresis can be easily observed in concentrated sunlight. Many different kinds of particles were tested. Some particles moved toward the sun and some moved away no matter which direction the sunlight was introduced into the chamber when the pressure of the air was from 0.5 to 30 torr or higher. Most particles moved along straight paths, but helical motions were also observed occasionally. In the convergent part of the beam, the particles moved toward the focus in the direction of the ray. Their speed increased gradually as they moved toward the focus, reached a maximum at the focus, and then slowed. It is very easy to observe photophoresis (particles moving either toward or away from the light source) at the focus when the beam is hot enough to burn paper. There is no apparent difference in the magnitude of the repelling force



and the attracting force of a concentrated sunlight beam in regions of similar light intensity.

A "funnel" effect was observed when the sunlight was introduced into the chamber from above. Carbon particles exhibited this best due to their low density and good photophoresis reaction. The particles in the beam were pushed toward the focus by the photophoretic force, while the particles in the reflected light outside the beam itself settled much slower. The particles at the boundary of the light beam moved along the boundary without falling from the beam. The phenomenon is very similar to the motion of particles in a liquid passing through a funnel. In this case the funnel wall is only the boundary between the light beam and the surrounding region. A force analysis for this case is shown in Figure 6. The direction of the photophoretic force is apparently in the direction of the wave front normal of the light. From Figure 6 it is easily seen that the net force,  $F_n$ , should push the particles out of the beam. Since this is not the case, there must exist another counteracting force which pulls the particles and keeps them in the beam. This phenomenon is like gravito-photophoresis, where the particles move up or down to the boundary in horizontal opposing beams and stay there without falling out. This phenomenon has not been interpreted in terms of the radiometer effect. A magnetizing effect of light might be considered here. The funnel

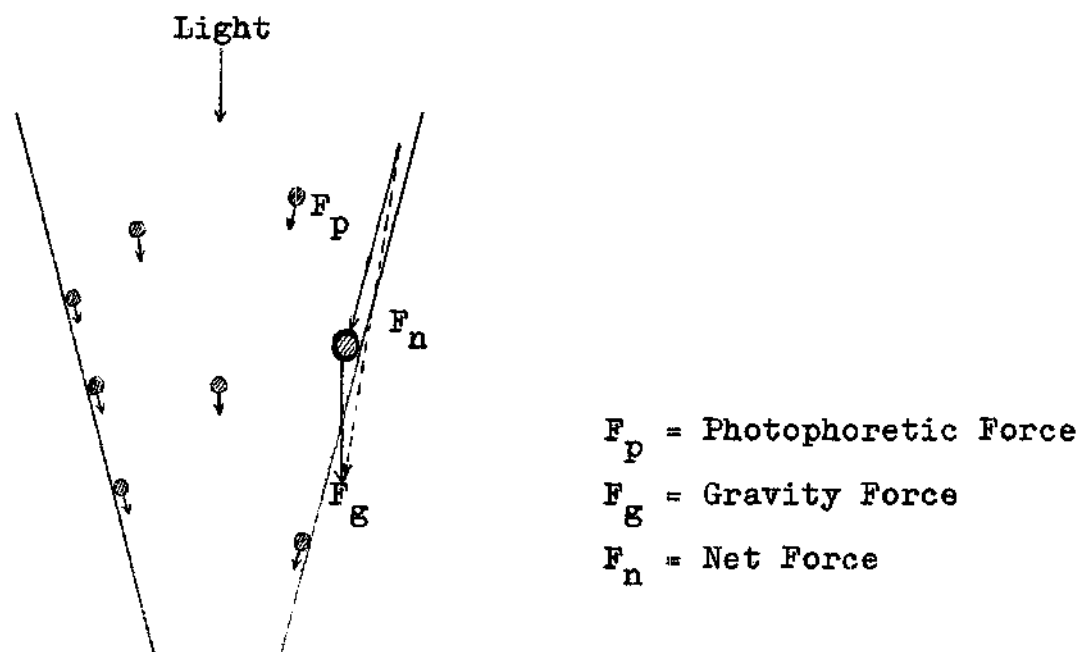


Figure 6. Force Analysis for Particle at the Edge of a Convergent Beam when the Light is from the Top

phenomenon happens only in a very high intensity converging light beam. If the intensity of the converging light beam is weak, the particles will pass through the boundary of the beam. The vertical elliptical motion described in the last section is also an example of what happens in a weak beam.

#### Parallel Beams

##### Stable Photophoresis

A nearly parallel beam of light was obtained by using the point-sources of light as described in a previous

section. In order to attain high intensities, a small diameter parallel beam of 1/2 inch diameter was used. The intensity was not entirely uniform across the beam, but regions having average energy densities of 0.185, 0.25, and 0.91 watt/cm<sup>2</sup> were used to measure forces and observe photophoretic phenomena.

The particles used in this investigation were not spheres. However, most of the photophoretic phenomena were very stable and free from irregular motion or particle rotation. The rising and falling velocities of particles in light and darkness could be measured with considerable reproducibility since straight paths were usually found for the particles. Some particles showed rotation and helical motion. Straight-path motion is due to the uniform forces acting on the particle while unbalanced forces cause particle rotation and helical or irregular trajectories.

#### Unstable Photophoresis

While most particles did exhibit stable reactions, those that did not are worthy of brief mention. Some particles were observed to start moving after being illuminated for a time. Some of them then moved in helical or irregular orbits. Some of them changed their directions very often, always moving parallel to the beam. Some of them occasionally lost their effect in light and fell. These phenomena were observed in a parallel light beam,

especially under pressures lower than 10 torr.

Under pressures of 2 torr, some particles were accelerated by the light to speeds of several inches per second. This included both light-positive and light-negative motion. Here irregular and unstable photophoresis phenomena commonly occur.

#### Rising and Falling Velocities

In order to calculate photophoretic forces and particle sizes, rising and falling velocities must be measured in vertical, parallel light beam of known intensity. The beam was introduced into the testing chamber from the top for light-negative measurements. Particles move toward the light source in the beam when the negative photophoretic force is greater than the gravity force and the particles fall due to the gravity force when the beam is interrupted. The beam was introduced into the chamber from the bottom for light-positive measurement. The particles move up and away from the light source when the positive photophoretic force is greater than the gravity force and fall due to the gravity force when the beam is off. The timer (see Chapter III, Shutter and Timer) could be adjusted to control the shutter so that reasonably stable rising and falling velocities could be obtained. The data are presented in Tables 14 through 20 in the Appendix.

There are two cases which could not be measured accurately by the method mentioned above. One was when

high speed motion was developed. This usually was encountered at gas pressures lower than 10 torr. The particles simply moved away before a measurement could be taken. The other was with very small particles of low density in gas pressures higher than 10 torr. Here settling was so slow that convection currents became significant.

The gases, air and helium, were utilized in testing the dependence of the photophoretic force on the medium. The properties of air and helium are compared in Table 4. Carbon particles were used. No apparent differences were observed in photophoretic phenomena under several pressures from 1 torr to atmospheric pressure. Both positive and negative motion was observed. The photophoretic velocity was measured at three pressures: 1.5, 10, and 30 torr. Similar velocities were obtained under these pressures, but at a pressure of 10 torr, photophoretic velocities measured in helium were beginning to be larger than those measured in air for particle radii larger than 0.5 micron. These data are given in Tables 18 and 19 in the Appendix.

#### Electric and Magnetic Field Effects

Observation of photophoretic phenomena in the presence of electric and magnetic fields further reveals the nature of photophoresis. An electric field was applied to a vertical parallel beam as shown in Figure 7. The potential gradient could be varied from zero to 10,000 volts/inch.

Table 4. Comparison of the Properties of Air and Helium

<u>Property</u>	<u>Pressure (Torr)</u>	<u>Air</u>	<u>Helium</u>	<u>References</u>
Absolute viscosity (Micro-poise) at 20° C and 1 atm.		181.94	196.14	(33) (34)
Mean Free Path (micron) at 20° C	1.5 10 30	33.9 5.1 1.7	96.7 14.5 4.8	by equation (21)
Mean Molecular Speed (cm/sec)		$4.65 \times 10^4$	$12.55 \times 10^4$	by equation (22)
Falling Velocity (cm/sec) due to gravity for a carbon particle with one micron radius	1.5 10 30	1.00 0.162 0.064	2.75 0.42 0.152	by equations (16) and (19)
Slip factor for a carbon particle with one micron radius	1.5 10 30	72.0 9.0 3.6	158.0 24.5 8.5	by equation (19)

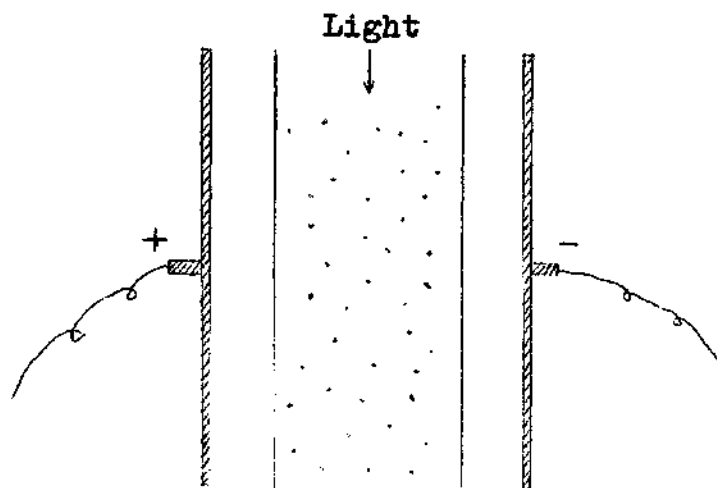


Figure 7. Photophoresis and Electric Field

Carbon particles were suspended in the cell as before. When illuminated by a vertical, parallel beam of light, some travelled toward the light source and some away from it, also as before. When the electric field was applied, however, particle behavior was altered, as summarized by the following statements: (1) Light-positive particles continued to move away from the light source and light-negative particles continued toward the light source. (2) Some particles turned to one electrode while others turned to the other, disregarding their behavior before the electric field was applied. (3) Some light-positive and some light-negative particles were accelerated (both positively and negatively) by the electric field. (4) Some particles changed from light positive to light negative or vice versa in the electric field.

Photophoresis in a vertical convergent-divergent beam was also examined in the presence of a magnetic field.

Carbon and iron particles were tested. The behavior of carbon particles in convergent and divergent beams has been described previously. Phenomena observed in a magnetic field were as follows:

(1) Of two particles in the convergent portion of the beam, one was stationary about  $1/8$  inch below another one which was moving in a  $1/16$ -inch-diameter circular orbit. When a strong magnet was brought near them as shown in Figure 8, the particle undergoing the circular motion moved higher by  $1/2$  inch while the other was unaffected. When the poles were reversed, the particle in the circular path was forced down about  $3/8$  inch below the other one, which stayed still. The field thus influenced only the one in motion.

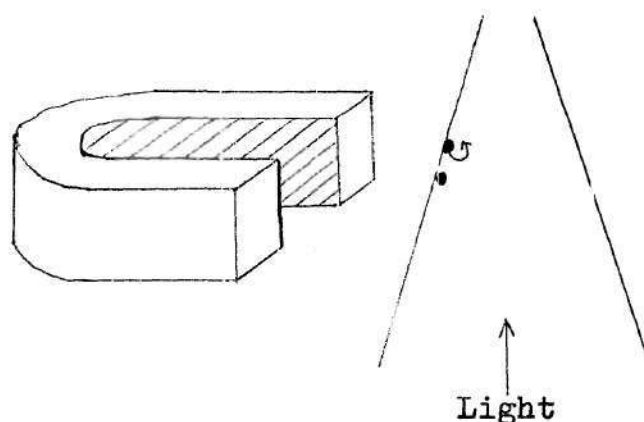


Figure 8. Photophoresis and Magnetic Field

(2) Another particle which rotated very rapidly about its own axis while retaining a fixed position in a converging beam was slowed and finally stopped when a strong



magnet was brought near. With the magnet near, the particle moved upward smoothly and slowly. When the magnet was taken away, the particle dropped again to its original position and started rotating again. The complete series phenomena was repeated many times.

(3) Still another was moving in about a 1/16-inch-diameter circular path near the edge of a convergent beam. When a magnet was brought near, the particle was driven from the beam entirely.

(4) A particle moving in a horizontal circular orbit in the divergent portion of a beam above the beam focus had its orbit enlarged until the particle moved out of the beam when a magnet was brought near. This phenomenon, too, has been observed many times.

(5) Some particles were not influenced at all by the magnet.

Iron particles were influenced by even a weak magnet. The phenomena observed were different each time. A few examples of the effect of a small magnet are presented below:

(1) The particles moving across a converging beam dropped out as shown in Figure 9 when the magnet was brought near.

(2) As the magnet approached the beam, some particles moved toward it and some moved away from it. Also, some particles were uninfluenced.

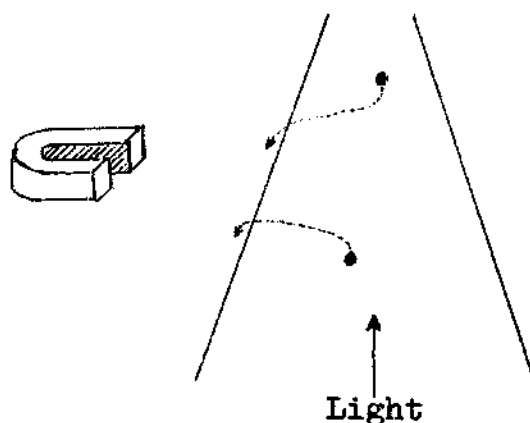


Figure 9. Iron Particle Crossing the Beam  
in Magnetic Field

(3) A particle was observed to move upward toward the focus when the magnet was near and to fall in the beam when the magnet was taken away. This phenomenon was noted on several occasions.

(4) Light-positive or negative particles could be accelerated (either positively or negatively) by the magnet.

(5) Particles moving in horizontal circular or vertical elliptical orbits were driven out of the beam by the magnet.

#### Wavelength Influence

In attempts to determine whether or not the ionization of gas molecules was involved in photophoresis, tests with several light filters using both sunlight and artificial light were made. The filters used are listed in Table 5.

Filter Nos. 1 through 5 were used in the test with light from the concentrated zirconium arc lamp and the

Table 5. Filters Used in Filter Test and Their Conditions

Filter No.	Per Cent of Wave Length ( $\text{\AA}$ ) Filtered Out				
	14 (%)	30 (%)	50 (%)	80 (%)	100 (%)
1	3800	3650	3590	3550	3450
2	5100	4420	4100	3920	3700
3	5900	5490	5410	5390	5200
4	6100	5760	5680	5660	5500
5	6500	6200	6150	6120	5950
6	6950	6500	6350	6310	6150

mercury arc lamp. Filter No. 6 cut out so much of the light that the particles in the beam could not be seen by an observer. The filters were used singly or in combination. Photophoresis still existed in all cases even though the photophoretic velocity was reduced considerably with each decrease in light intensity.

Filter Nos. 1 through 6 were also tested using sunlight. Photophoresis again existed in all cases. Another heat absorbing glass which permits only light with wave length from 4,000 to 10,000  $\text{\AA}$  was tested in this instance. Although heat energy was reduced drastically, photophoresis was still observable.

Since gas molecules illuminated by these filtered beams of light cannot be ionized, photophoresis must not depend on gas molecule ionization.

### Analysis of Errors

Scientific data inevitably involve errors as a result of mathematical simplifications, empirical equations, and experimental measurements. The errors in this work are those of method and technique.

#### Method Errors

The method of evaluation of particle size was based on the assumption that all particles are spheres. This is not true although it appears that most of the particles do approximate spherical shape. An empirical equation was used to calculate the slip factor which is also based on the assumption that all particles are spheres. This equation disregards any dependence on the nature of particles, such as with the accommodation coefficient. Since there was a very small difference between particle and gas molecule temperature, the empirical equation for slip factor was probably acceptable. The assumptions introduced in the calculation of the mean free path of the gas molecules were that the viscosity of the gas is constant and that the gas nature is ideal under low pressures. These are more than likely satisfactory assumptions.

In the calculation of the gravity force on particles, the bulk density was used except for wood charcoal. Some particles, especially those of carbon, gas carbon, and zinc, were agglomerated. The errors in assessing particle density and particle size were probably rather large for

these agglomerated particles. The errors would be compensating to some extent since they would tend to indicate increased particle sizes and decreased particle densities.

#### Experimental Errors

The velocities of rise and fall of the particles were measured by a vertical displacement apparatus and recorded. Some velocities were read directly from a scale on the window of the chamber. Data for several trips up and down for each particle deviated by no more than 10 per cent for the information to be used. The errors due to timer, shutter, and recorder were less than 2 per cent. Air convection effects at pressures higher than 30 torr must be considered so the measurements were restricted to lower values.

Pressures were measured by an absolute pressure gage of Wallace and Tierman, Belleville, New Jersey. Pressure variation during a test was maintained within 0.5 torr or lower. Room temperature was fairly constant at about 22° C, but a temperature nearer 25° C existed for the air in the chamber during a test. The beam was not completely parallel, and its intensity distribution was not entirely uniform. All velocities were measured in one region of the beam, however, so the data are consistent.

From the discussion it is evident that the most significant errors in the evaluation of photophoretic forces arise from uncertainties in particle properties such as size and density. These errors varied with individual

particles due to the condition of agglomeration. The errors of method and the systematic experimental inaccuracies are inherently identical for all cases. Overall, the uncertainty of silicon carbide and sodium chloride particles is probably on the order of  $\pm 50$  per cent. Errors for other particles are somewhat higher.

## CHAPTER V

### DISCUSSION OF RESULTS

#### Comparison of Positive and Negative Photophoresis

Both positive and negative photophoretic forces for sodium chloride and silicon carbide particles at several different pressures are compared in Figures 24, 26, 27, and Figures 30 through 33 in the Appendix. The results are different for different particles and different pressures. For sodium chloride particles positive photophoretic forces are almost identical with the negative photophoretic ones for the same size particle under 50 torr pressure. Positive photophoretic forces are considerably larger than the negative photophoretic forces at 15 torr pressure. At 30 torr pressure, negative photophoretic forces are larger than the positive ones only for particles less than one micron in radius. For silicon carbide particles, the positive photophoretic forces are larger than the negative at pressures of 5, 30, and 50 torr, while at 10 torr pressure the negative photophoretic forces are slightly larger than the positive ones for particles less than one micron in radius. Overall, most of the positive photophoretic forces are of the same order of magnitude as the negative forces.

### Dependence on Gas Medium

No differences were observed between positive and negative photophoretic forces in air and in helium, despite the differences in the physical properties of air and helium under a pressure of 10 torr. For pressure of 10 torr, the photophoretic forces measured in helium are greater than those measured in air for particles of radius larger than 0.5 micron. This difference may be due to errors in applying the slip factor equation (19) to helium gas at that pressure. There is not sufficient information on this in the literature to permit a definite conclusion. These calculated data are presented in Tables 18 and 19 and Figures 35 through 37 in the Appendix, and Figures 10 through 12. Parankiewicz<sup>(44)</sup> gave only three measurements with selenium particles at around atmospheric pressure in argon, nitrogen, and hydrogen gases. The similarity of photophoretic forces is shown in Table 6. These data were taken by exposing selenium particles to a horizontal beam of light. The photophoretic speed means the horizontal component of the particle speed which was caused by the photophoretic force alone.

### Dependence on the Light Intensity

Photophoretic force is very strongly dependent on the light intensity. Two different intensity lamps were used. One produced  $0.185 \text{ watt/cm}^2$  and the other one  $0.25 \text{ watt/cm}^2$  of energy, the latter being about 35 per cent



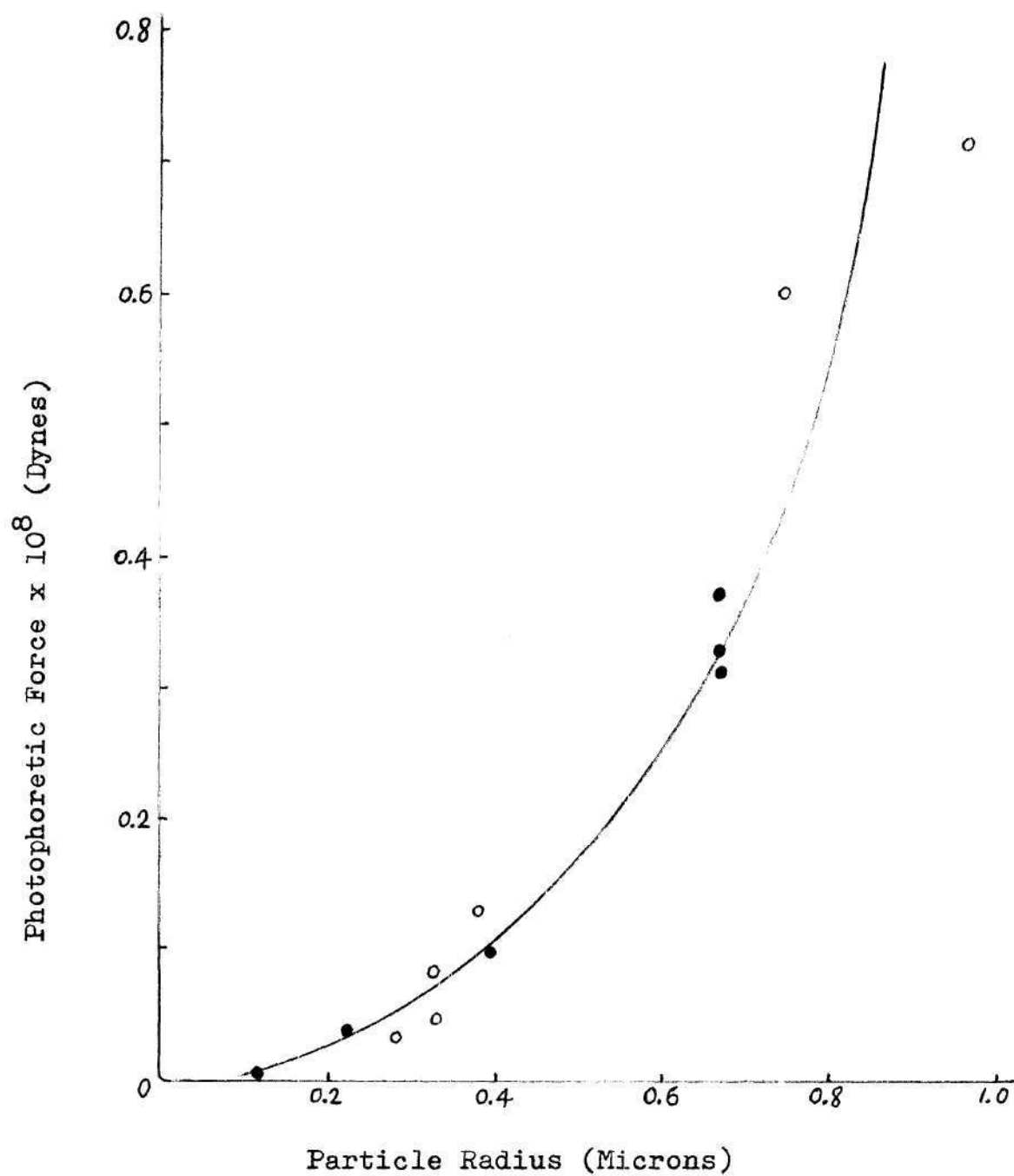


Figure 10. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 1.5 Torr in Helium (•) and Air (o).

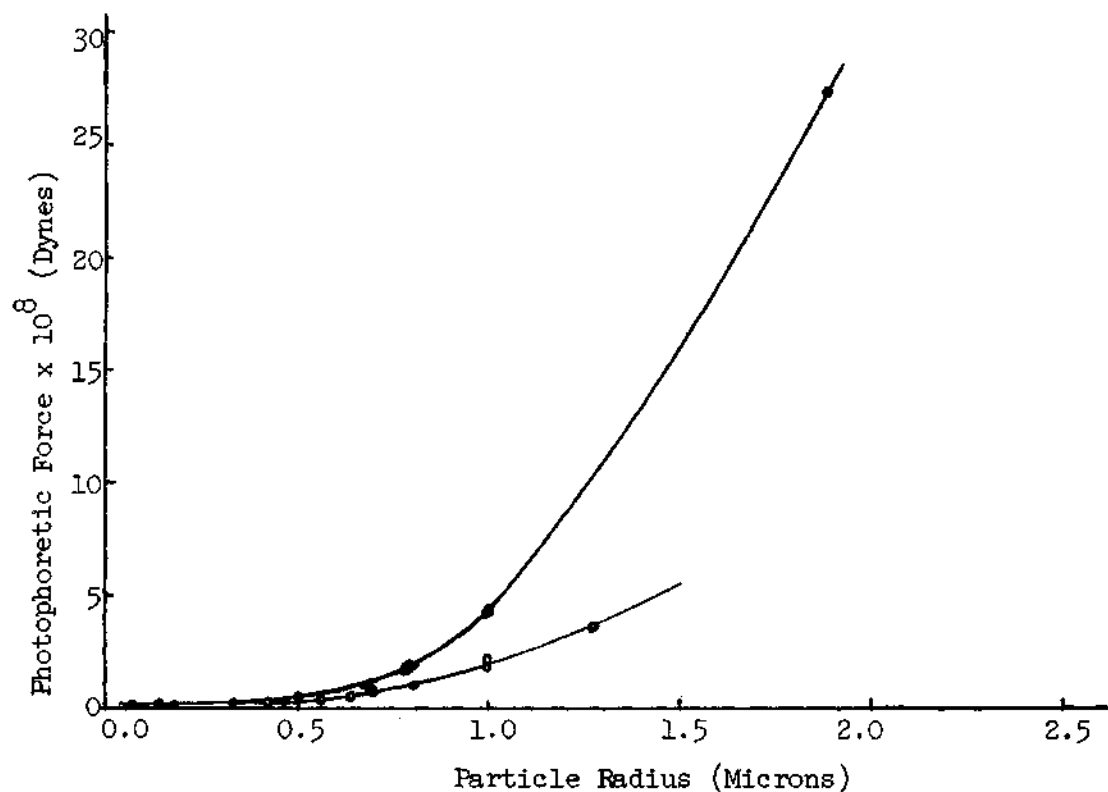


Figure 11. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 10 Torr in Helium (●) and Air (○).

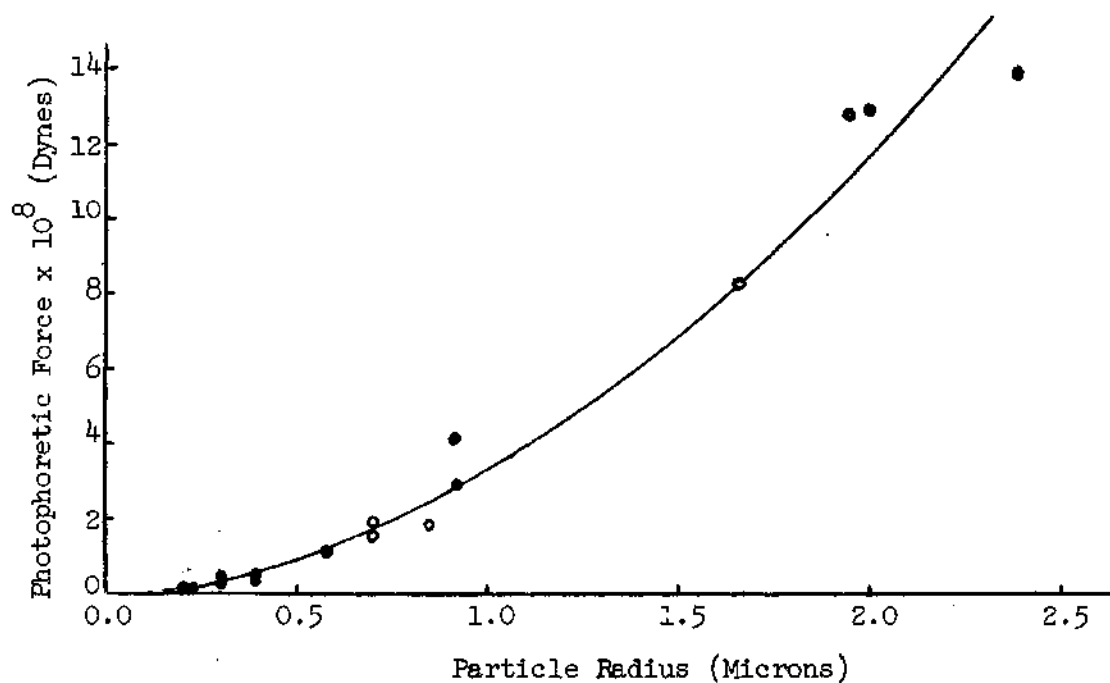


Figure 12. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 30 Torr in Helium (●) and Air (○).

Table 6. Photophoretic Forces of Selenium Particles in Argon, Nitrogen, and Hydrogen Measured by Parankeiwicz

Gas	Falling Speed (cm/sec)	Radius (micron)	Photophoretic Speed (cm/sec)	Photophoretic Force (dyne)
Ar	$1.53 \times 10^{-3}$	0.1530	$11.72 \times 10^{-3}$	$4.85 \times 10^{-10}$
N <sub>2</sub>	$1.91 \times 10^{-3}$	0.1526	$14.37 \times 10^{-3}$	$4.70 \times 10^{-10}$
H <sub>2</sub>	$5.16 \times 10^{-3}$	0.1515	$38.68 \times 10^{-3}$	$4.78 \times 10^{-10}$

stronger than the former. A comparison of the results with gas carbon particles at 25 torr pressure shows that the forces measured in the  $0.25 \text{ watt/cm}^2$  light is about 30 to 40 per cent stronger than in the  $0.185 \text{ watt/cm}^2$  light. This suggests that the photophoretic force is directly proportional to the intensity of the light. These data are compared in Figure 13 taken from the thesis of Rosen.<sup>(50)</sup>

#### Effect of Particle Differences

Different particles in an intense beam of light give different effects. Some show strong photophoresis, some show weak photophoresis, while some do not reveal photophoresis at all. Even for the same kind of material, particles may show positive photophoresis, negative photophoresis, irregular photophoresis, or no photophoresis. Therefore, it must be that photophoresis depends strongly

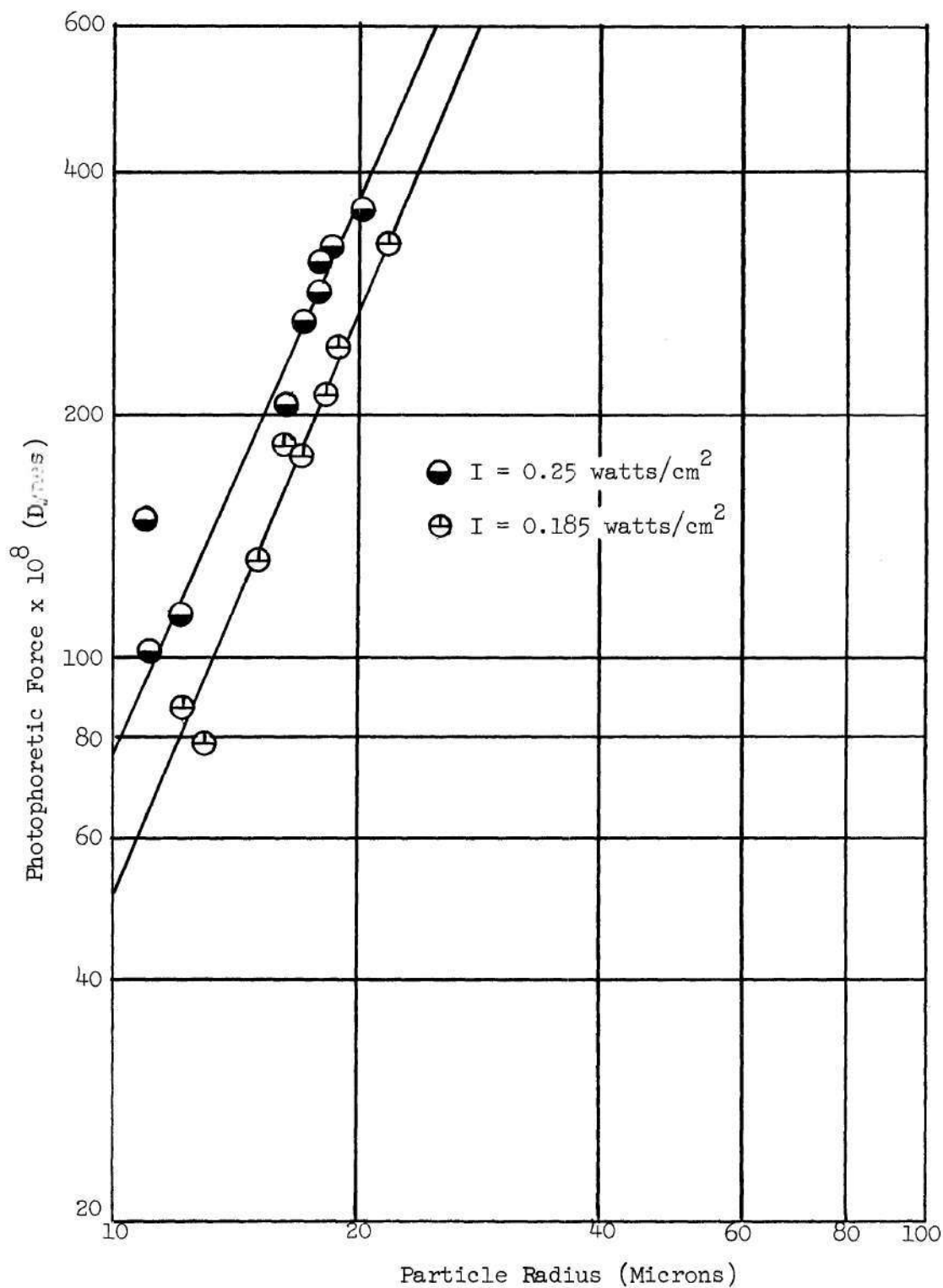


Figure 13. Comparison of Positive Photophoretic Force for Gas Carbon Particles at a Pressure of 25 Torr Taken at Different Light Intensities ( $I$ ).

on the nature of individual particles, perhaps their size, shape, thermal conductivity, agglomeration, light reflection and refraction, heat absorption, heat capacity, electric charge, etc. All these characteristics except charges are considered in the following discussion.

Photophoretic force depends strongly on the particle size. A representative plot of photophoretic force versus particle radius for zinc particles at 10 torr is shown in Figure 14. According to equation (4), if it is assumed that the accommodation coefficient equals 0.8, then the approximate value for  $P_{\max}$  of each particle of radius,  $a$ , is as follows:

$a$ (micron)	0.1	0.5	1.0	2.0	5.0	10	20	30
$P_{\max}$ (torr)	1340	268	134	67	26.8	13.4	6.7	4.5

For particles less than 2 microns in radius, photophoretic forces measured in the pressure region below 67 torr should follow the low pressure equation equation (7) if the radiometer point of view is correct. The force is proportional to  $a^2 \Delta T$  where  $\Delta T$  is a function of particle size, the thermal conductivity of particle and gas, and the intensity and heat energy of the light. If conditions are kept constant, the force will be proportional to some power of the radius larger than 2 and depend on the relation between  $\Delta T$  and  $a$ .

From equations (6), (7), and (8), it may be con-

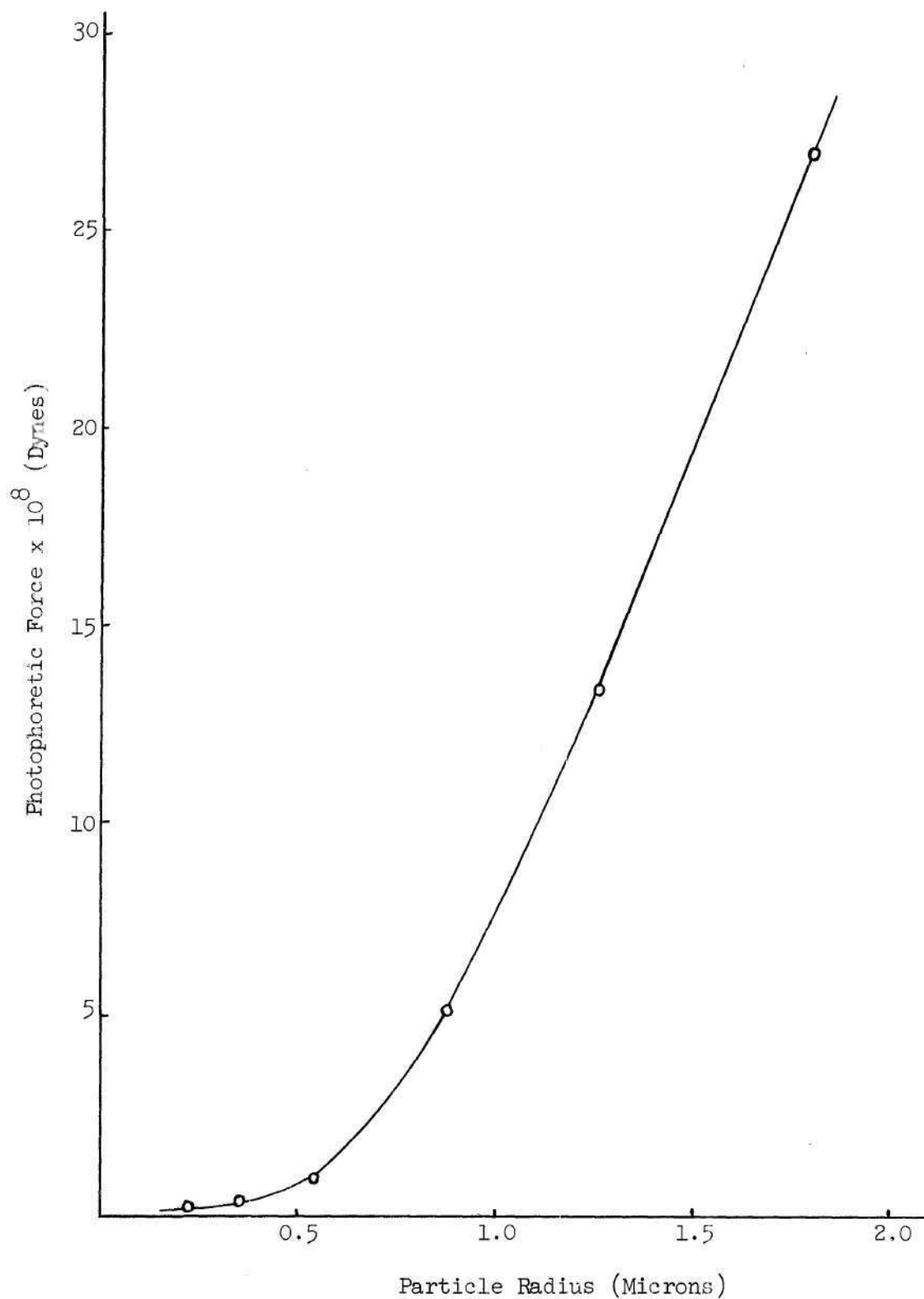


Figure 14. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 10 Torr.

cluded that the higher (relatively) the pressure, the less the dependence of the force on the radius. Most of the data were measured in the low pressure region with a few in the transition and high pressure regions. The exponents of the radius term are determined from log-log plots of photophoretic force versus radius. These exponents vary from 1.70 to 3.57. These data are presented in Table 7 and Figure 15.

Table 7. Exponent on Particle Radius Term  
as a Function of Gas Pressure

1. Positive Photophoretic Force of Sodium Chloride Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
0.33 - 1.81	15	2.45
0.44 - 1.25	20	2.41
0.75 - 2.04	30	2.45
0.59 - 2.41	50	2.69
0.77 - 1.28	63	3.19

2. Negative Photophoretic Force of Sodium Chloride Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
0.15 - 1.80	10	2.50
0.33 - 2.22	15	1.76
0.22 - 2.48	30	1.70
0.34 - 1.15	50	2.69

Table 7. Exponent on Particle Radius Term  
as a Function of Gas Pressure (Continued)

3. Positive Photophoretic Force of Silicon Carbide Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
0.14 - 0.72	2	2.91
0.17 - 0.98	5	2.64
0.40 - 2.15	10	2.72
0.54 - 1.15	30	3.26
0.42 - 1.45	50	2.84

4. Negative Photophoretic Force of Silicon Carbide Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
0.85 - 2.60	5	3.57
0.40 - 0.95	10	2.96
0.29 - 1.15	30	1.78
0.42 - 0.75	50	2.20

5. Positive Photophoretic Force of Zinc Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
0.13 - 1.10	4	3.06
0.23 - 1.81	10	2.83
0.26 - 1.42	20	3.11
0.12 - 2.07	30	2.63

(Continued)



Table 7. Exponent on Particle Radius Term  
as a Function of Gas Pressure (Concluded)

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6. Negative Photophoretic Force of Carbon Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
0.40 - 0.79	0.6	2.70
0.28 - 0.95	1.5	2.46
0.14 - 1.27	10	2.64
0.20 - 2.39	30	2.47

7. Positive Photophoretic Force of Wood Charcoal Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
6.56 - 12.8	4.7	2.85
6.92 - 12.5	9.7	2.80
2.22 - 7.9	15.6	2.33
2.44 - 9.7	21.3	3.00

8. Positive Photophoretic Force of Gas Carbon Particles

<u>Particle Radius Range</u> (microns)	<u>Pressure</u> (torr)	<u>Exponent</u>
17.4 - 30.0	8.3	2.72
14.0 - 24.1	15.3	2.78
11.0 - 20.3	25.6	2.30
8.4 - 22.5	35.4	1.86
14.2 - 19.5	46	1.80

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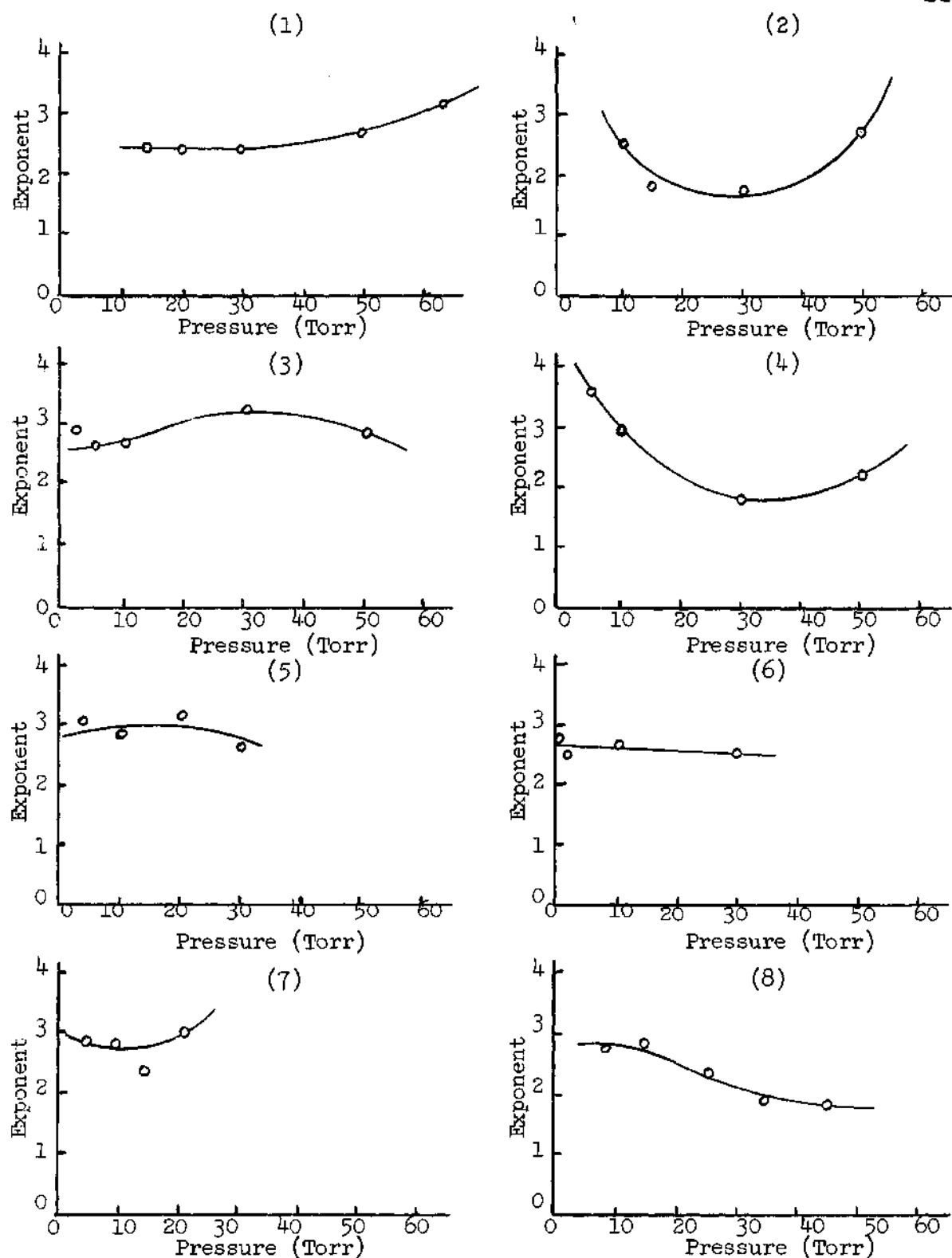


Figure 15. Exponent on Particle Radius Term versus Gas Pressure as Shown in Table VI.

In the low pressure region, particle radii were found to be in the range from 0.1 to 2.6 microns. For positive photophoresis, the exponent of radius does not vary much for silicon carbide and zinc particles. But for sodium chloride particles, the exponent is constant below about 30 torr pressure and increases with pressures over 30 torr. For negative photophoresis, a minimum is found around pressures of 30 torr for sodium chloride and silicon carbide particles.

In the transition pressure region, particle radii were from 2 to 13 microns according to Rosen.<sup>(50)</sup> An average value of 2.80 was obtained for the wood charcoal particles. In the high pressure region, the exponent of radius decreases with increasing pressure. The particles used were gas carbon with radii from 11 to 30 microns.

The dependence of photophoretic force on particle material can be observed very easily because some materials show very strong photophoresis and others do not.

Some of the calculated values of the photophoretic force for different materials are shown in Table 8 and Figures 16 and 17. The positive photophoretic force of sodium chloride particles is considerably smaller than it is for zinc and silicon carbide particles. But the negative photophoretic force for sodium chloride particles is much larger than it is for the carbon and silicon carbide particles. The results give strong support to the inter-

Table 8. A Comparison of Photophoretic Force for Different Materials at 30 Torr

<u>Material</u>	<u>Particle Radius</u> (micron)	<u>Photophoretic Force</u> (dyne x 10 <sup>-8</sup> )
Positive Photophoresis		
NaCl	0.75	2.41
SiC	0.76	3.70
Zn	0.71	3.25
NaCl	1.54	10.255
	(1.15)	(6.0)(Figure 16)
SiC	1.15	13.93
NaCl	2.04	25.64
Zn	2.07	60.80
Negative Photophoresis		
NaCl	0.42	1.37
SiC	0.42	0.61
C	0.39	0.55
NaCl	2.48	24.52
C	2.39	13.90

pretation of photophoresis as a radiometer effect, for, from the discussion of the radiometer effect given in Chapter II, Radiometer Effect, it may be seen that the positive photophoretic force for translucent particles should be smaller than for nontranslucent particles, and the negative photophoretic force of translucent particles should be larger than for nontranslucent ones.

The positive photophoretic force of zinc particles is larger than it is for sodium chloride particles even

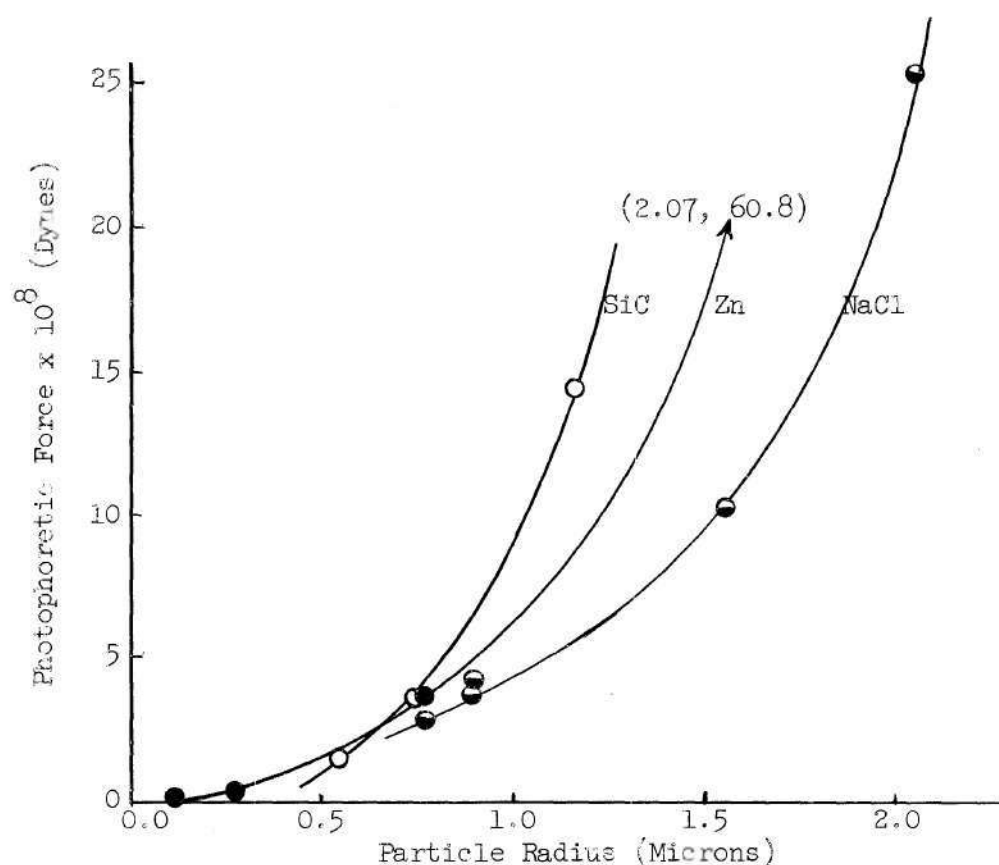


Figure 16. Positive Photophoretic Force versus Particle Radius for Sodium Chloride (●), Silicon Carbide (○), and Zinc (●) Particles at a Pressure of 30 Torr.

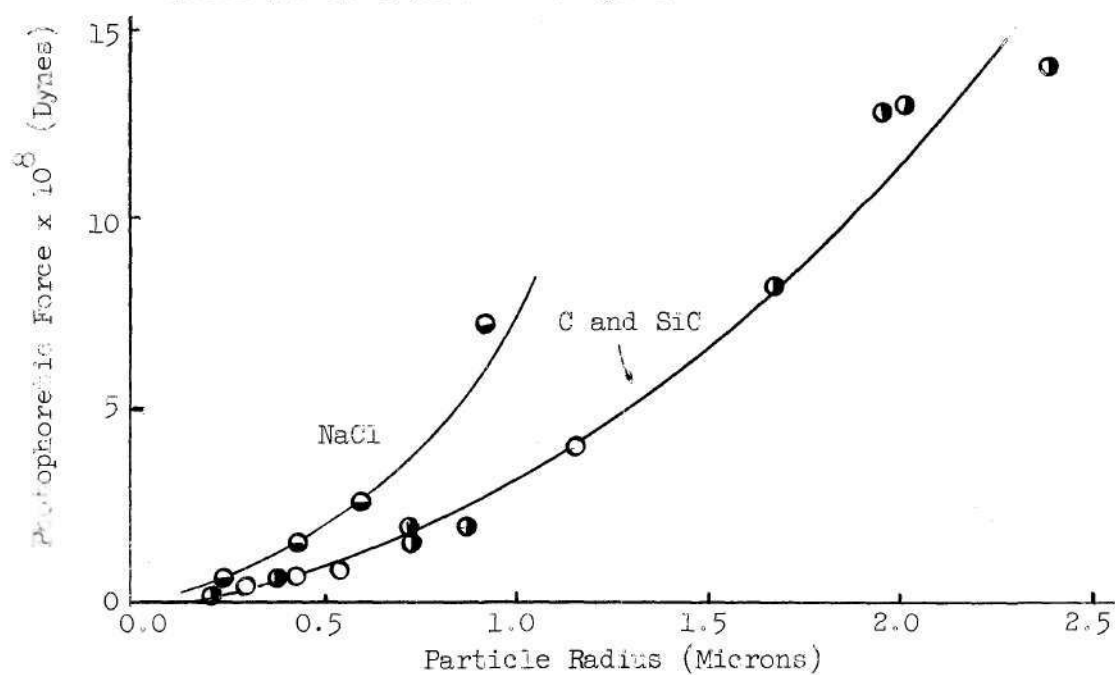


Figure 17. Negative Photophoretic Force versus Particle Radius for Carbon (●), Sodium Chloride (●), and Silicon Carbide (○) Particles at a Pressure of 30 Torr.

though the thermal conductivity of zinc is much higher than that of sodium chloride. This indicates that the thermal conductivity is not very important for such small particles. The negative photophoretic force on sodium chloride particles is much larger than for carbon particles even though the radiation energy absorption of carbon particles should be much better than for sodium chloride particles. This indicates that the temperature distribution on the particle surface is more important than the mean temperature of the particle.

The reports of Parankiewicz<sup>(44)</sup> and Ehrenhaft<sup>(12)</sup> (13) show that photophoretic forces vary very little in the pressure range from 55 to 760 torr. Several particles from 0.09 to 0.23 micron in radius were studied by these investigators. In contradiction to this, Mattauch<sup>(37)</sup>(39) found that the photophoretic force was like a radiometer force and depended on the gas pressure.

From observation of photophoretic phenomena, it is apparent that the photophoretic force is a function of pressure. Since unavoidable convection currents disturb the measurement of photophoretic force at high pressures, reliable measurements could only be obtained at low pressures. This makes it difficult to study the dependence of photophoretic force on gas pressure. The experimental results fail to give a clear overall conclusion about photophoretic force and gas pressure. Some results show

that a maximum photophoretic force does exist at a certain pressure. Good examples of this are given in Table 9 and Figures 18, 19, 20, and 21.

Table 9. Photophoretic Force as a Function of Pressure of the Gaseous Medium

1. Negative Photophoretic Force of 0.38 micron radius sodium chloride particle

<u>Pressure</u> (torr)	<u>Photophoretic Force</u> (dyne x 10 <sup>-8</sup> )	<u>Note</u>
2	0.075	Measured Value
10	0.31	Measured Value
15	0.60	Smoothed Value
30	1.30	Smoothed Value
50	0.60	Smoothed Value

2. Positive Photophoretic Force of 0.72 micron radius silicon carbide particle

<u>Pressure</u> (torr)	<u>Photophoretic Force</u> (dyne x 10 <sup>-8</sup> )	<u>Note</u>
2	1.13	Measured Value
5	2.1	Smoothed Value
10	1.4	Smoothed Value
30	3.4	Smoothed Value
50	3.0	Smoothed Value

3. Negative Photophoretic Force of 0.50 micron radius carbon particle

<u>Pressure</u> (torr)	<u>Photophoretic Force</u> (dyne x 10 <sup>-8</sup> )	<u>Note</u>
0.6	0.19	Smoothed Value
1.5	0.20	Smoothed Value
10	0.40	Smoothed Value
30	0.64	Smoothed Value

4. Negative Photophoretic Force of 0.90 radius sodium chloride particle

<u>Pressure</u> (torr)	<u>Photophoretic Force</u> (dyne x 10 <sup>-8</sup> )	<u>Note</u>
10	2.72	Measured Value
15	2.7	Smoothed Value
30	5.0	Smoothed Value
50	5.6	Smoothed Value



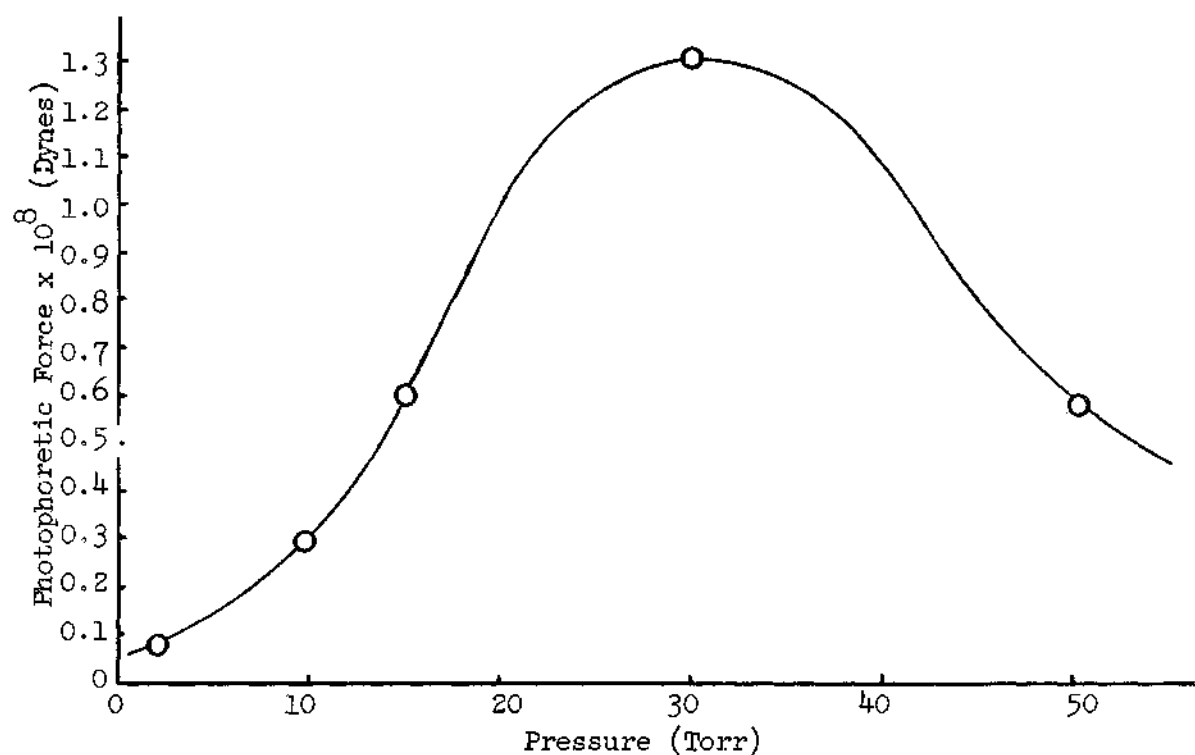


Figure 18. Negative Photophoretic Force versus Pressure for 0.38 Micron Radius Silicon Carbide Particle.

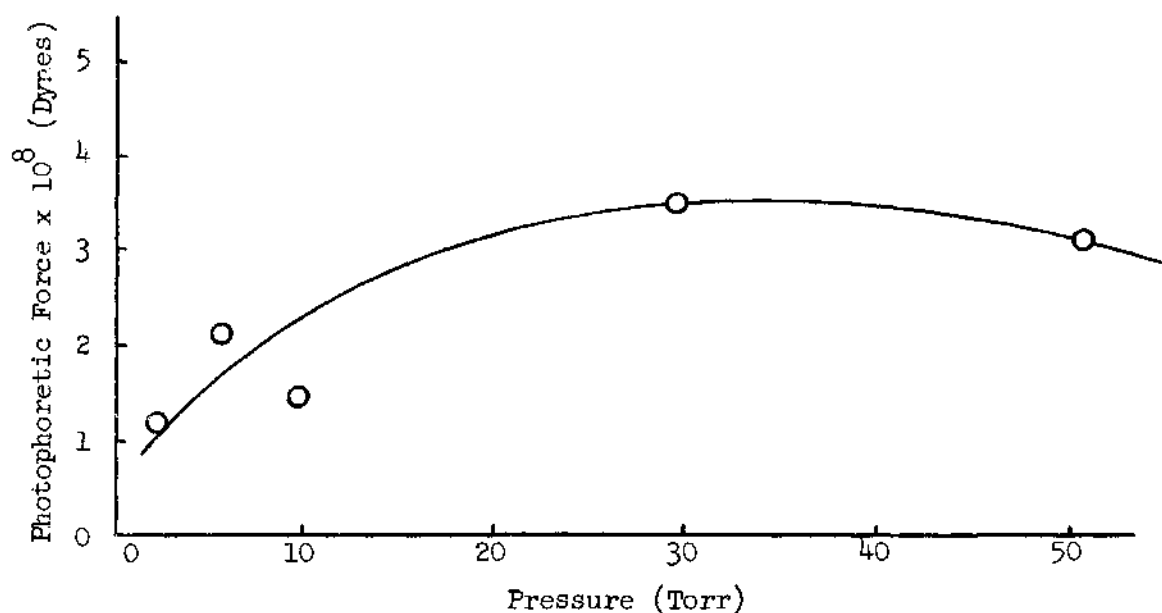


Figure 19. Positive Photophoretic Force versus Pressure for 0.72 Micron Radius Silicon Carbide Particle

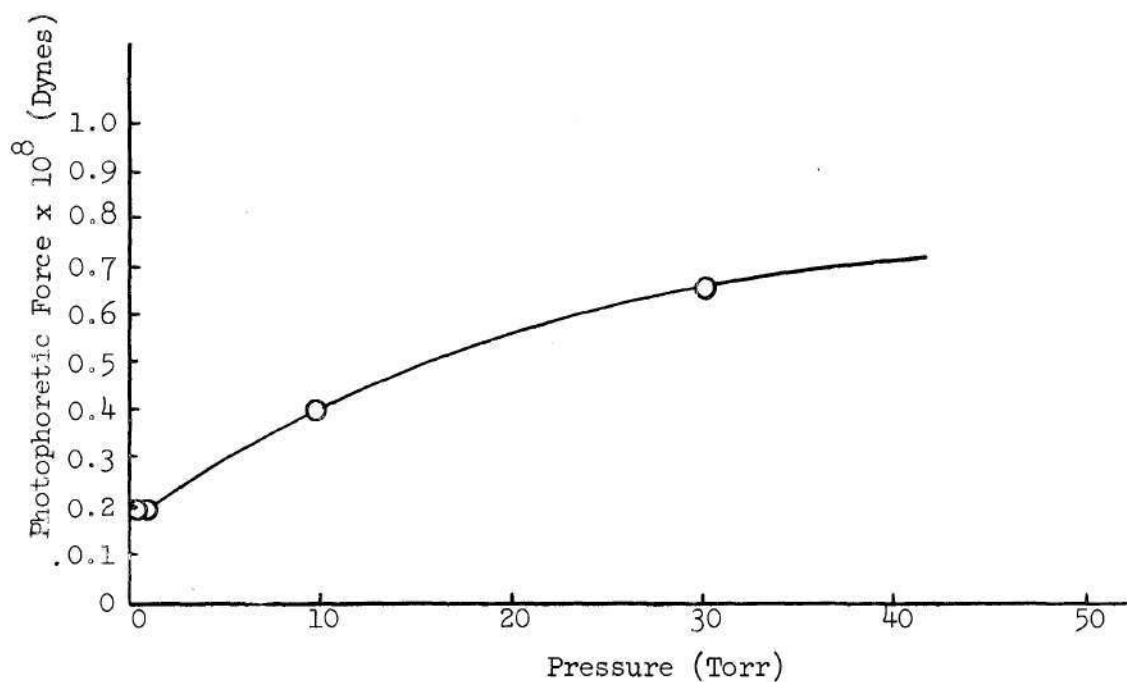


Figure 20. Negative Photophoretic Force versus Pressure for 0.5 Micron Radius Carbon Particle.

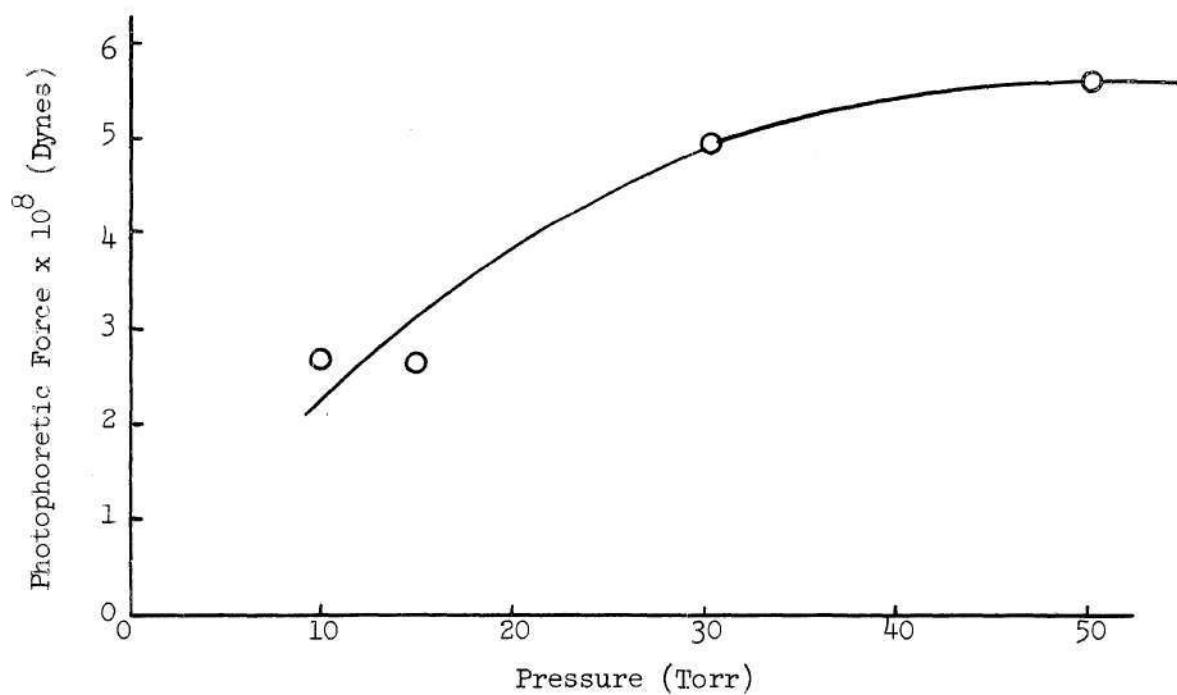


Figure 21. Negative Photophoretic Force versus Pressure for 0.9 Micron Radius Sodium Chloride Particles.

## CHAPTER VI

### RELEVANCE TO METEOROLOGICAL PHENOMENA

Photophoresis may be of some importance at high altitudes. Very small particles exposed to sunlight at high altitudes where the pressure is naturally low will undoubtedly experience a photophoretic force. If negative, the particles will be carried to higher levels or, if positive, driven to lower ones.

#### Substances in High Altitudes

Much effort has been expended on high altitude sampling in recent years. Balloons, aircraft, and rockets have been employed. From the reports on these stratospheric aerosol studies,<sup>(7)(31)</sup> the particle concentration profiles show a maximum around an altitude of 12 miles in a band there that is about 5 miles thick. The particles range from less than 0.1 micron in radius to 2.0 microns radius. Their analysis indicates that sulfur is the most important element, and that it is most likely present in the form of sulfate. It thus appears that the sulfates of ammonium and sodium are most likely to be the particles present in the stratosphere. Junge and Manson<sup>(31)</sup> find that particles with 1 micron radius in a column of 6 miles will be removed in about half a year if they are assumed to be 1 micron in

radius and to have a density of  $2 \text{ gm/cm}^3$  and fall with a velocity of  $0.07 \text{ cm/sec}$ . This should then be the approximate maximum lifetime of these particles.

Only a few particles in the stratosphere containing sulfate have radii greater than 1.5 microns. Stebbins<sup>(54)</sup> reports that most of these large particles have a higher electron optical density and more irregular outlines than do sulfate particles. Occasionally, spheres of high density with diameters smaller than 1 micron were found. Some of these nonsulfate particles may be extraterrestrial in origin. Radioactive particles due to the tests of nuclear weapons have also been studied in the High Altitude Sampling Program<sup>(54)</sup>. The particles contain strontium<sup>-90</sup>, tungsten<sup>-185</sup>, barium<sup>-140</sup>, strontium<sup>-89</sup>, cerium<sup>-144</sup>, rhodium<sup>-102</sup>, and plutonium. Natural radioactivity in the upper air includes carbon<sup>-14</sup>, tritium, beryllium<sup>-7</sup>, phosphorus<sup>-32</sup>, and lead<sup>-210</sup>. Tungsten particles were found to show photophoresis in an intense beam of light in this study.

Sodium was observed in the twilight spectrum at about 85 Km (53 miles) altitude in the form of ionized atoms.<sup>(30)</sup> Very fine liquid droplets from the ocean surface may be caught up by the wind and carried by air convection upward in the form of sodium chloride crystals. In this study sodium chloride particles were observed that showed very strong negative photophoresis. In the strato-

sphere, hydrogen sulfide and sulfur dioxide can be oxidized by ozone or intense ultraviolet light<sup>(7)(30)(31)</sup> to form sulfates. Sodium and ammonium are the possible cations of the sulfates.<sup>(30)(31)</sup>

Soberman<sup>(53)</sup> has studied noctilucent clouds which occur 50 miles high during the summer at high latitudes. Samples of them were brought back by rocket-borne collectors in Sweden. It was found that the number of particles collected during the time when noctilucent clouds were evident was between 100 and 1,000 times more than on a cloudless night. The size of the particles mostly ranged from 0.05 to 0.5 micron in diameter. About 20 per cent of the particles from noctilucent clouds were coated by ice. The solid particles were found to contain nickel, an element quite rare in terrestrial particles but common in those of meteoritic origin. Nickel particles were observed here that showed photophoresis when exposed to an intense beam of light. No work has been done on the photophoresis of ice-coated particles up to now.

#### Estimation of Photophoresis of Atmospheric Particles

Experimental results (see Figure 13) show that the photophoretic force is linearly proportional to the intensity of light at constant pressure. According to the observation reported here, ammonium sulfate, sodium sulfate, and sodium chloride particles show photophoretic phenomena when

exposed to an intense beam of light. So the study of sodium chloride will give a rough idea of how particles are influenced by photophoresis in the stratosphere. The region of maximum particle concentration is from 10 to 15 miles altitude where the pressures are from about 75 to 40 torr. Smoothed photophoretic forces on sodium chloride particles at pressures of 30 and 50 torr (which are equivalent to 17 and 14 miles altitude respectively) and their extrapolations to the intensity of sunlight in the stratosphere are given in Tables 10 and 11.

The intensity of sunlight in the stratosphere is estimated to be 85 per cent of the solar constant which is given as  $430 \text{ Btu/hr-ft}^2(6)$  or  $0.135 \text{ watt/cm}^2$ . The intensity of sunlight measured on the earth's surface is about 70 per cent of the solar constant due to the absorption and reflection of the atmosphere about the earth. Particles caught in the stratosphere have a density of approximately  $2.0 \text{ gm/cm}^3$ . (The density of  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  is  $1.982 \text{ gm/cm}^3$ , that of  $(\text{NH}_4)_2\text{SO}_4$  is  $1.769 \text{ gm/cm}^3$ , and that of  $\text{NaCl}$  is  $2.165 \text{ gm/cm}^3$ .) Calculated positive and negative photophoretic forces, gravity forces, rising velocities for those light-negative particles and falling velocities for those light-positive particles when the sun is at meridian and settling velocities in darkness are given in Tables 12 and 13, assuming the pressure to be 30 and 50 torr.

As shown in Tables 12 and 13, it is calculated that

Table 10. Estimated Values of Negative Photophoretic Forces in Stratosphere

Particle Radius of Sodium Chloride Particles (micron)	Photophoretic Force at Intensity = 0.91 watt/cm <sup>2</sup> (Mercury arc lamp) (dyne)	Extrapolation Value of Photophoretic Force at Intensity = 0.108 watt/cm <sup>2</sup> (dyne)
Pressure = 30 torr		
0.1	$1.5 \times 10^{-9}$	$1.78 \times 10^{-10}$
0.2	$4.0 \times 10^{-9}$	$4.75 \times 10^{-10}$
0.5	$2.0 \times 10^{-8}$	$2.37 \times 10^{-9}$
1.0	$6.4 \times 10^{-8}$	$7.60 \times 10^{-9}$
2.0	$2.0 \times 10^{-7}$	$2.37 \times 10^{-8}$
Pressure = 50 torr		
0.1	$1.6 \times 10^{-10}$	$1.90 \times 10^{-11}$
0.2	$9.5 \times 10^{-10}$	$1.13 \times 10^{-10}$
0.5	$1.2 \times 10^{-8}$	$1.43 \times 10^{-9}$
1.0	$7.2 \times 10^{-8}$	$8.57 \times 10^{-9}$
2.0	$4.8 \times 10^{-7}$	$5.71 \times 10^{-8}$

Table 11. Estimated Values of Positive Photophoretic Forces in Stratosphere

Particle Radius of Sodium Chloride Particles (micron)	Photophoretic Force at Intensity = 0.91 watt/cm <sup>2</sup> (Mercury arc lamp) (dyne)	Extrapolation Value of Photophoretic Force at Intensity = 0.108 watt/cm <sup>2</sup> (dyne)
Pressure = 30 torr		
0.1	$1.7 \times 10^{-10}$	$2.16 \times 10^{-11}$
0.2	$8.1 \times 10^{-10}$	$9.61 \times 10^{-11}$
0.5	$8.0 \times 10^{-9}$	$9.50 \times 10^{-10}$
1.0	$4.6 \times 10^{-8}$	$5.46 \times 10^{-9}$
2.0	$2.4 \times 10^{-7}$	$2.85 \times 10^{-8}$
Pressure = 50 torr		
0.1	$1.6 \times 10^{-10}$	$1.90 \times 10^{-11}$
0.2	$9.5 \times 10^{-10}$	$1.13 \times 10^{-10}$
0.5	$1.2 \times 10^{-8}$	$1.43 \times 10^{-9}$
1.0	$7.2 \times 10^{-8}$	$8.57 \times 10^{-9}$
2.0	$4.8 \times 10^{-7}$	$5.71 \times 10^{-8}$



Table 12. Estimated Rising and Settling Velocities of the Light-Negative Particles in the Stratosphere

a	$F_p$	$F_g$	$V_r = \frac{(F_p - F_g) S}{6 \pi \mu a}$	$V_s = \frac{F_g S}{6 \pi \mu a}$
Particle Radius (micron)	Photophoretic Force (dyne)	Gravity Force (dyne)	Rising Velocity (cm/sec)	Settling Velocity (cm/sec)
Pressure = 30 torr				
0.1	$1.78 \times 10^{-10}$	$8.21 \times 10^{-12}$	$+ 1.34 \times 10^{-1}$	$6.49 \times 10^{-3}$
0.2	$4.75 \times 10^{-10}$	$6.57 \times 10^{-11}$	$+ 8.35 \times 10^{-2}$	$1.34 \times 10^{-2}$
0.5	$2.37 \times 10^{-9}$	$1.03 \times 10^{-9}$	$+ 4.69 \times 10^{-2}$	$3.60 \times 10^{-2}$
1.0	$7.60 \times 10^{-9}$	$8.21 \times 10^{-9}$	$- 6.13 \times 10^{-3}$	$8.24 \times 10^{-2}$
2.0	$2.37 \times 10^{-8}$	$6.57 \times 10^{-8}$	$- 1.35 \times 10^{-1}$	$2.12 \times 10^{-1}$
Pressure = 50 torr				
0.1	$1.90 \times 10^{-11}$	$8.21 \times 10^{-12}$	$+ 5.41 \times 10^{-3}$	$4.12 \times 10^{-3}$
0.2	$1.13 \times 10^{-10}$	$6.57 \times 10^{-11}$	$+ 6.20 \times 10^{-3}$	$8.61 \times 10^{-3}$
0.5	$1.43 \times 10^{-9}$	$1.03 \times 10^{-9}$	$+ 9.56 \times 10^{-3}$	$2.46 \times 10^{-2}$
1.0	$8.57 \times 10^{-9}$	$8.21 \times 10^{-9}$	$+ 2.62 \times 10^{-3}$	$5.98 \times 10^{-2}$
2.0	$5.71 \times 10^{-8}$	$6.57 \times 10^{-8}$	$- 2.22 \times 10^{-2}$	$1.70 \times 10^{-1}$

Table 13. Estimated Falling Velocities of the Light-Positive Particles in the Stratosphere

$a$ Particle Radius (micron)	$F_p$ Photophoretic Force (dyne)	$F_g$ Gravity Force (dyne)	$V_F = \frac{(F_p - F_g) S}{6 \pi \mu a}$ Falling Velocity (cm/sec)	$V_s = \frac{F_g S}{6 \pi \mu a}$ Settling Velocity (cm/sec)
Pressure = 30 torr				
0.1	$2.16 \times 10^{-11}$	$8.21 \times 10^{-12}$	$2.35 \times 10^{-2}$	$6.49 \times 10^{-3}$
0.2	$9.61 \times 10^{-11}$	$6.57 \times 10^{-11}$	$3.31 \times 10^{-2}$	$1.34 \times 10^{-2}$
0.5	$9.50 \times 10^{-10}$	$1.03 \times 10^{-9}$	$6.94 \times 10^{-2}$	$3.60 \times 10^{-2}$
1.0	$5.46 \times 10^{-9}$	$8.21 \times 10^{-9}$	$1.38 \times 10^{-1}$	$8.24 \times 10^{-2}$
2.0	$2.85 \times 10^{-8}$	$6.57 \times 10^{-8}$	$3.05 \times 10^{-1}$	$2.12 \times 10^{-1}$
Pressure = 50 torr				
0.1	$1.90 \times 10^{-11}$	$8.21 \times 10^{-11}$	$1.34 \times 10^{-2}$	$4.12 \times 10^{-3}$
0.2	$1.13 \times 10^{-10}$	$6.57 \times 10^{-11}$	$2.31 \times 10^{-2}$	$8.61 \times 10^{-3}$
0.5	$1.43 \times 10^{-9}$	$1.03 \times 10^{-9}$	$5.80 \times 10^{-2}$	$2.46 \times 10^{-2}$
1.0	$8.57 \times 10^{-9}$	$8.21 \times 10^{-9}$	$1.21 \times 10^{-1}$	$5.98 \times 10^{-2}$
2.0	$5.71 \times 10^{-8}$	$6.57 \times 10^{-8}$	$3.13 \times 10^{-1}$	$1.70 \times 10^{-1}$

some particles smaller than 1 micron radius will rise in sunlight against the gravity force and some particles larger than 1 to 1.5 microns radius will fall more slowly in sunlight than they would under the influence of gravity alone. Those particles with a positive photophoresis will fall more than twice as fast in the sunlight as in the dark for most cases. It seems likely, therefore, that some particles in the stratosphere will not be replaced in the period of time formerly predicted. Some particles less than 0.1 micron, which rise in the sunlight and fall in the dark, may even stay within a certain region indefinitely unless air convection brings them down. Settling velocities for particles with a density of  $2.0 \text{ gm/cm}^3$  are given versus altitude<sup>(30)</sup> on Figure 22. Banet<sup>(2)</sup> and Ehrenhaft<sup>(21)</sup> had tried to relate photophoresis to the earth's magnetism and the gravitation of celestial bodies. This is too far beyond present knowledge of photophoresis to be indulged in here.

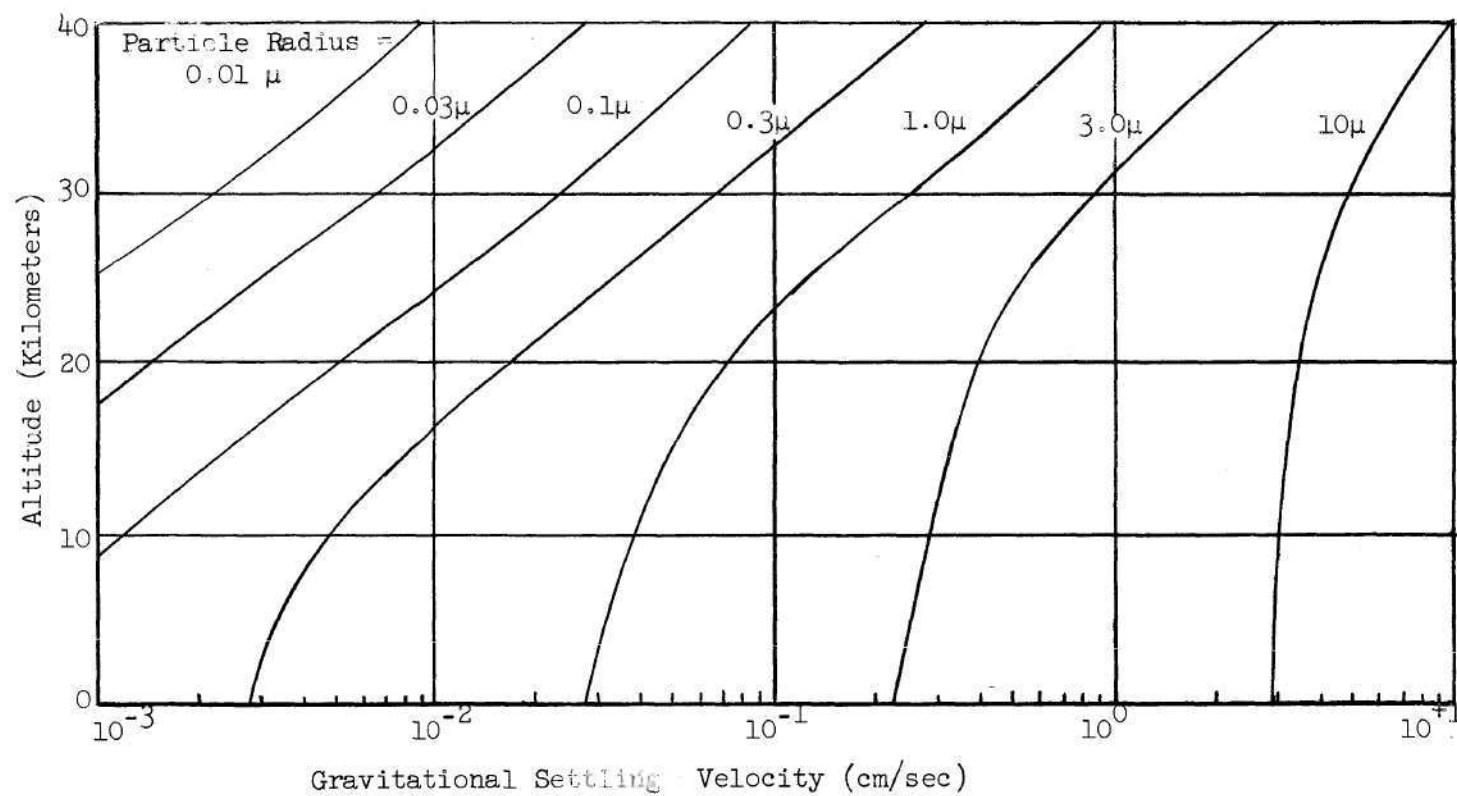


Figure 22. Falling Velocities for Particles with a Density of  $2.0 \text{ gm/cm}^3$  versus Altitude<sup>(30)</sup>

## CHAPTER VII

### FINAL REMARKS

According to the observations and measurements made here, a satisfactory theory of photophoresis must account for a number of phenomena. Among them are:

#### Straight Path in Light's Direction

When irregularly shaped particles are exposed in a parallel beam of light, some of them move while others do not. Among the moving particles, only two directions are usually seen--toward or away from the light source. According to the radiometer effect, the force should act against the particle surface where the temperature is higher than the rest. Since the net force acting on a particle is in the direction of the parallel beam regardless of the shape of the particle, this indicates that there may exist lines of force in the direction of the light which resist the particles crossing them. The same nature of light beam appears at the boundary, too, but boundary phenomena will be discussed in a later section.

#### Changing Directions

Particles were observed changing direction in both parallel and nonparallel light beams. This happens most

often under pressures lower than 10 torr. Particles move along a straight path parallel to the direction of the ray, turning back and forth with regularity. Ehrenhaft<sup>(19)</sup> thought that the light could induce charges on particles and this certainly could change them. It is hard to explain, however, how light changes the charges or why it does so on particles at low pressures predominantly. Particles also rotate along the axis parallel to the ray in a beam of light.<sup>(25)(47)</sup> From the radiometer point of view, particles move faster under low pressures and may rotate with their axis perpendicular to the direction of the light ray. Each revolution then will coincide with a complete oscillation. The experimental observations support this explanation because all the particles which change directions do move rapidly. Particles are not likely to change their directions if they move slowly. The curved motion in a nonparallel beam can also be interpreted by the radiometer effect due to the rotation of particles, but no way to interpret it by electric and magnetic charges has yet been offered.

#### Initiation of Photophoresis

Particles apparently start to move as soon as exposed to light and stop as soon as the light is covered. If the electric and magnetic charge explanation is accepted, there is no problem in interpreting this phenomenon because

the beam of light itself is considered to be behaving as a stationary electric and magnet field.<sup>(17)</sup> This can also be interpreted by the radiometer effect. Because they are so small, the particles will almost immediately be heated on one side so the radiometer force would act on them as soon as the light is uncovered.

#### Boundary Phenomena

Phenomena of the boundary can hardly be explained by the radiometer effect without considering the boundary of a beam to behave as a net of magnetic force lines. Otherwise, the particle cannot stay at the beam boundary, and it should be driven out by the radiometer force.

#### Influence of Electric and Magnetic Fields

Some particles were influenced by electric or magnetic fields and some were not. Results suggest that the phenomena of photophoresis in the electric or magnetic field are combinations of photophoresis and field effect. There is no evidence that light-positive particles and light-negative particles carry different charges.

#### Positive and Negative Photophoretic Forces

From the radiometer point of view, opaque particles should have larger positive photophoretic forces and smaller negative photophoretic forces than would translucent particles. Very small particles are all translucent so

they may show light negative motion, but large particles should be opaque and not show negative photophoresis. The experimental observations and measurements, at least, do not conflict with this.

#### Dependence on Particle Material and Gas

Particles of the same material and same size do not necessarily show the same phenomenon even in the same position within a light beam. Therefore, photophoresis must depend on the condition of the individual particle, such as its temperature distribution, charges, etc., no matter what is the state of the gas. The photophoretic force depends on the pressure of the gas, so photophoresis must not be only a matter between light and particles but one also with the gaseous medium. If the pressure of the gas does not influence the electric and magnetic ions induced on particles by light as Ehrenhaft considered,<sup>(19)</sup> then interpretation by electric and magnetic ions will not be sufficient to describe the dependence of the photophoretic force on gas pressure. It is, of course, unlikely that light, especially after passing a filter (see Table 5), can still induce charges on particles.



## CHAPTER VIII

### CONCLUSIONS

In view of the foregoing, it is concluded that:

1. In both parallel and nonparallel beams of light, with electric and magnetic fields, or without any field save that of gravity, the radiometer effect affords part of the explanation for photophoresis. There must be other interactions, however, existing between a small particle and a beam of light, for a heating effect does not permit a complete description of all phenomena.

2. Because photophoresis exists in long wave length beams of light (  $6000 \text{ \AA}$  ), ionization of gas molecules alone cannot explain photophoresis.

3. Photophoresis is only slightly dependent on the nature of the gaseous medium. In many cases it is essentially independent of the gas.

4. Individual particles of all the materials tested (radii from 0.1 to 2.0 microns) showed both positive and negative photophoresis in intense light. The largest light-negative particle measured was only 2.60 microns in radius while the largest light-positive particle was 30.0 microns. This result gives strong support to the interpretation of photophoresis as a radiometer effect.

5. Particles in the stratosphere are most likely both pulled upward and pushed downward by the photophoretic action of sunlight. Some particles with radii less than 0.1 micron may even be pulled upward farther when in the sunlight than they are pulled downward in the dark by gravity.

6. Because photophoresis exists to quite low pressures, the fine particles may be made to rise to high altitudes in the atmosphere.

## CHAPTER IX

### RECOMMENDATIONS

Additional studies should be made under conditions similar to those actually existing at high altitudes. A high-vacuum, temperature-regulated system is needed to determine at what altitude photophoretic phenomena end. For example, a vacuum of  $10^{-2}$  torr is equivalent to 50 miles in altitude where the ambient temperature is about  $-80^{\circ}$  C, a vacuum of  $10^{-6}$  torr is equivalent to 100 miles altitude where the ambient temperature is about  $260^{\circ}$  C, and a vacuum of  $10^{-7}$  torr is equivalent to 140 miles altitude where the effective temperature is approximately  $500^{\circ}$  C.<sup>(29)</sup> The apparatus should also be one in which particles can be suspended for long times while exposed to sunlight. The chamber should be long enough to test photophoresis as a function of time to see if there is any relation with time of exposure. Ice crystals particularly should be examined for their photophoretic response. Other particles which are thought to exist at high altitude, both natural and from nuclear tests, ought also to be considered. Special apparatus and techniques must be devised to test ice-coated particles and humidified particles which have been found in noctilucent clouds.

In order to study whether there are any relationships among photophoresis, the earth's magnetism, and gravity, a study on larger solid bodies suspended in a constant, intense light source for a long period is necessary. A simple test might be made by suspending a ball by means of a thread in a chamber free from outside interferences. An intense beam of light should then be introduced into the chamber to shine on the ball. A window surrounding the chamber is required so that the position of the ball can be seen from any direction. A graphite ball is suggested for use due to its low density and good photophoretic response. Marks should be inscribed on the ball so that any rotation of it could be noted.

A more intense light is needed to test the reaction of larger particles than was employed in this study. Whether there is any maximum and minimum particle size for light-positive and light-negative particles would be of interest. This knowledge could have a bearing on the interpretation of photophoresis as a radiometer effect, and it might help to establish new ideas about photophoresis. More intense lighting might also permit determining whether or not there is a saturation intensity with photophoresis as there is with magnetic fields, for example.

The relation between particle properties and photophoresis could be studied by using coated particles. The difference in photophoretic force and phenomena among

coated particles, particles of the coat material, and the particles of the inner materials might elucidate the dependence of photophoresis on particle properties.

Further studies on the light wave length and the nature of the gaseous medium are also needed. Intensely hot and "cool" lights should be compared for the photophoretic reaction they can produce.

## APPENDIX\*

\*All the data presented here were taken in the region where the light intensity was  $0.91 \text{ watt/cm}^2$ .

Table 14. Experimental Data of Positive Photophoretic Force for Sodium Chloride Particles

Falling Velocity (cm/sec)	Rising Velocity (cm/sec)	Particle Radius (microns)	Photophoretic Force (dynes x 10 <sup>-8</sup> )
Pressure = 15 torr			
0.048	0.681	0.33	0.49
0.080	0.795	0.54	1.55
0.318	1.133	1.81	25.91
Pressure = 20 torr			
0.048	0.272	0.44	0.50
0.095	0.340	0.80	2.08
0.112	0.295	0.92	2.47
0.159	0.340	1.25	5.43
Pressure = 30 torr			
0.064	0.340	0.75	2.41
0.080	0.340	0.91	3.54
0.080	0.386	0.91	3.93
0.159	0.340	1.54	10.26
0.239	0.566	2.04	25.64
Pressure = 50 torr			
0.032	0.226	0.59	1.49
0.064	0.566	1.00	8.77
0.795	0.566	1.15	11.20
0.239	1.133	2.41	71.83
Pressure = 63 torr			
0.040	0.226	0.77	2.65
0.064	0.340	1.09	7.28
0.080	0.340	1.28	9.80
0.080	0.566	1.28	15.08

Table 15. Experimental Data of Negative Photophoretic Force for Sodium Chloride Particles

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes x 10 <sup>-8</sup> )
Pressure = 2 torr			
0.396	0.160	0.38	0.075
Pressure = 10 torr			
0.064	0.160	0.15	0.018
0.127	0.160	0.27	0.066
0.159	0.254	0.33	0.15
0.175	0.318	0.38	0.31
0.396	0.635	0.90	2.72
0.795	0.254	1.80	8.24
Pressure = 15 torr			
0.048	0.572	0.33	0.43
0.064	0.635	0.43	0.81
0.095	0.254	0.62	0.81
0.159	0.381	1.01	3.25
0.175	0.318	1.10	3.42
0.414	0.032	2.22	10.44
Pressure = 30 torr			
0.016	0.794	0.22	0.48
0.032	0.635	0.42	1.37
0.048	0.635	0.60	2.74
0.079	0.794	0.91	7.38
0.318	0.254	2.48	24.52

(Continued)



Table 15. Experimental Data of Negative Photophoretic Force for Sodium Chloride Particles (Continued)

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes $\times 10^{-8}$ )
Pressure = 50 torr			
0.016	0.159	0.34	0.38
0.032	0.254	0.59	1.65
0.032	0.318	0.59	2.02
0.048	0.318	0.81	3.62
0.048	0.318	0.81	4.23
0.795	0.476	1.15	9.63

Table 16. Experimental Data of Positive Photophoretic Force for Silicon Carbide Particles

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes $\times 10^{-8}$ )
Pressure = 2 torr			
0.191	0.191	0.14	0.0068
0.318	0.445	0.21	0.029
0.448	0.635	0.28	0.10
1.113	1.430	0.72	1.13
Pressure = 5 torr			
0.095	0.381	0.17	0.030
0.095	0.445	0.17	0.034
0.318	1.270	0.43	0.62
0.318	1.430	0.43	0.68
0.635	1.905	0.98	4.85
Pressure = 10 torr			
0.127	0.318	0.40	0.30
0.160	0.318	0.50	0.49
0.191	0.381	0.60	0.85
0.795	0.953	2.15	28.97
Pressure = 30 torr			
0.064	0.381	0.54	1.46
0.095	0.508	0.76	3.70
0.160	0.953	1.15	13.93
Pressure = 50 torr			
0.032	0.191	0.42	0.70
0.064	0.318	0.75	3.32
0.127	0.478	1.22	11.62
0.160	0.795	1.45	24.07

Table 17. Experimental Data of Negative Photophoretic Force for Silicon Carbide Particles

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes x 10 <sup>-8</sup> )
Pressure = 5 torr			
0.556	0.159	0.85	1.05
0.635	0.159	0.98	1.54
0.953	0.476	1.48	6.42
1.748	2.540	2.60	56.52
Pressure = 10 torr			
0.127	0.318	0.40	0.30
0.159	0.318	0.50	0.49
0.191	0.794	0.60	1.46
0.238	0.635	0.72	2.01
0.318	0.699	0.95	3.65
Pressure = 30 torr			
0.032	0.254	0.29	0.29
0.048	0.254	0.42	0.61
0.064	0.159	0.54	0.73
0.159	0.159	1.15	4.00
Pressure = 50 torr			
0.032	0.095	0.42	0.40
0.048	0.127	0.60	1.04
0.048	0.191	0.60	1.42
0.064	0.127	0.75	1.66

Table 18. Experimental Data of Negative Photophoretic Force for Carbon Particles in Air

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes x 10 <sup>-8</sup> )
Pressure = 0.6 torr			
0.953	1.270	0.40	0.094
1.270	1.588	0.52	0.20
1.905	2.540	0.79	0.72
Pressure = 1.5 torr			
0.265	0.566	0.28	0.033
0.318	0.396	0.32	0.047
0.318	0.953	0.32	0.083
0.371	1.031	0.38	0.13
0.742	0.953	0.74	0.60
0.953	0.318	0.95	0.71
Pressure = 10 torr			
0.021	0.159	0.14	0.015
0.021	0.191	0.14	0.018
0.064	0.127	0.41	0.13
0.085	0.191	0.55	0.35
0.085	0.222	0.55	0.39
0.095	0.191	0.61	0.43
0.106	0.238	0.68	0.65
0.127	0.238	0.80	0.94
0.159	0.318	0.99	1.84
0.159	0.396	0.99	2.14
0.212	0.351	1.27	3.47

(Continued)

Table 18. Experimental Data of Negative Photophoretic Force for Carbon Particles in Air (Continued)

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes x 10 <sup>-8</sup> )
Pressure = 30 torr			
0.011	0.111	0.20	0.059
0.021	0.286	0.39	0.55
0.042	0.238	0.71	1.51
0.042	0.318	0.71	1.94
0.053	0.191	0.86	1.87
0.127	0.222	1.67	8.14
0.159	0.269	1.95	12.69
0.169	0.238	2.03	12.83
0.212	0.127	2.39	13.90

Table 19. Experimental Data of Negative Photophoretic Force for Carbon Particles in Helium

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes x 10 <sup>-8</sup> )
Pressure = 1.5 torr			
0.277	1.351	0.11	0.0048
0.589	2.474	0.22	0.035
1.176	0.826	0.46	0.10
1.765	1.237	0.66	0.31
1.765	1.443	0.66	0.33
1.765	1.854	0.66	0.37
Pressure = 10 torr			
0.027	0.266	0.07	0.0022
0.027	0.665	0.07	0.0053
0.039	0.375	0.10	0.0064
0.045	0.250	0.11	0.018
0.054	0.266	0.12	0.0071
0.060	0.266	0.14	0.0098
0.063	0.450	0.15	0.018
0.068	0.334	0.16	0.016
0.069	0.676	0.16	0.029
0.078	0.300	0.19	0.021
0.078	1.000	0.19	0.060
0.082	0.399	0.20	0.030
0.114	0.209	0.28	0.039
0.114	0.417	0.28	0.065
0.114	1.043	0.28	0.14
0.117	0.750	0.29	0.11
0.136	0.292	0.33	0.073

(Continued)

Table 19. Experimental Data of Negative Photophoretic Force for Carbon Particles in Helium (Continued)

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes $\times 10^{-8}$ )
Pressure = 10 torr			
0.138	0.676	0.34	0.13
0.152	0.676	0.38	0.18
0.163	1.011	0.41	0.30
0.182	0.417	0.45	0.19
0.182	0.626	0.45	0.25
0.207	1.013	0.50	0.47
0.326	1.729	0.78	1.89
0.414	2.568	0.98	4.32
0.828	4.392	1.89	27.21
Pressure = 30 torr			
0.028	0.338	0.20	0.066
0.041	0.338	0.30	0.16
0.041	0.541	0.30	0.25
0.055	0.406	0.39	0.32
0.055	0.607	0.39	0.45
0.083	0.676	0.57	1.09
0.138	0.676	0.91	2.90
0.138	1.013	0.91	4.11

Table 20. Experimental Data of Positive Photophoretic Force for Zinc Particles

<u>Falling Velocity</u> (cm/sec)	<u>Rising Velocity</u> (cm/sec)	<u>Particle Radius</u> (microns)	<u>Photophoretic Force</u> (dynes x 10 <sup>-8</sup> )
Pressure = 4 torr			
0.254	0.254	0.13	0.014
0.635	0.572	0.32	0.20
0.794	2.223	0.41	0.84
1.429	1.746	0.82	3.57
1.905	2.858	1.10	9.66
Pressure = 10 torr			
0.159	0.318	0.23	0.11
0.254	0.318	0.36	0.31
0.381	0.445	0.54	1.00
0.635	1.016	0.88	5.16
0.953	1.270	1.25	13.38
1.429	0.794	1.81	26.99
Pressure = 20 torr			
0.095	0.127	0.26	0.12
0.254	0.635	0.68	3.15
0.476	0.476	1.12	8.34
0.635	0.953	1.42	21.48
Pressure = 30 torr			
0.032	0.254	0.12	0.036
0.064	0.381	0.25	0.65
0.191	0.826	0.71	3.25
0.794	0.635	2.07	60.80



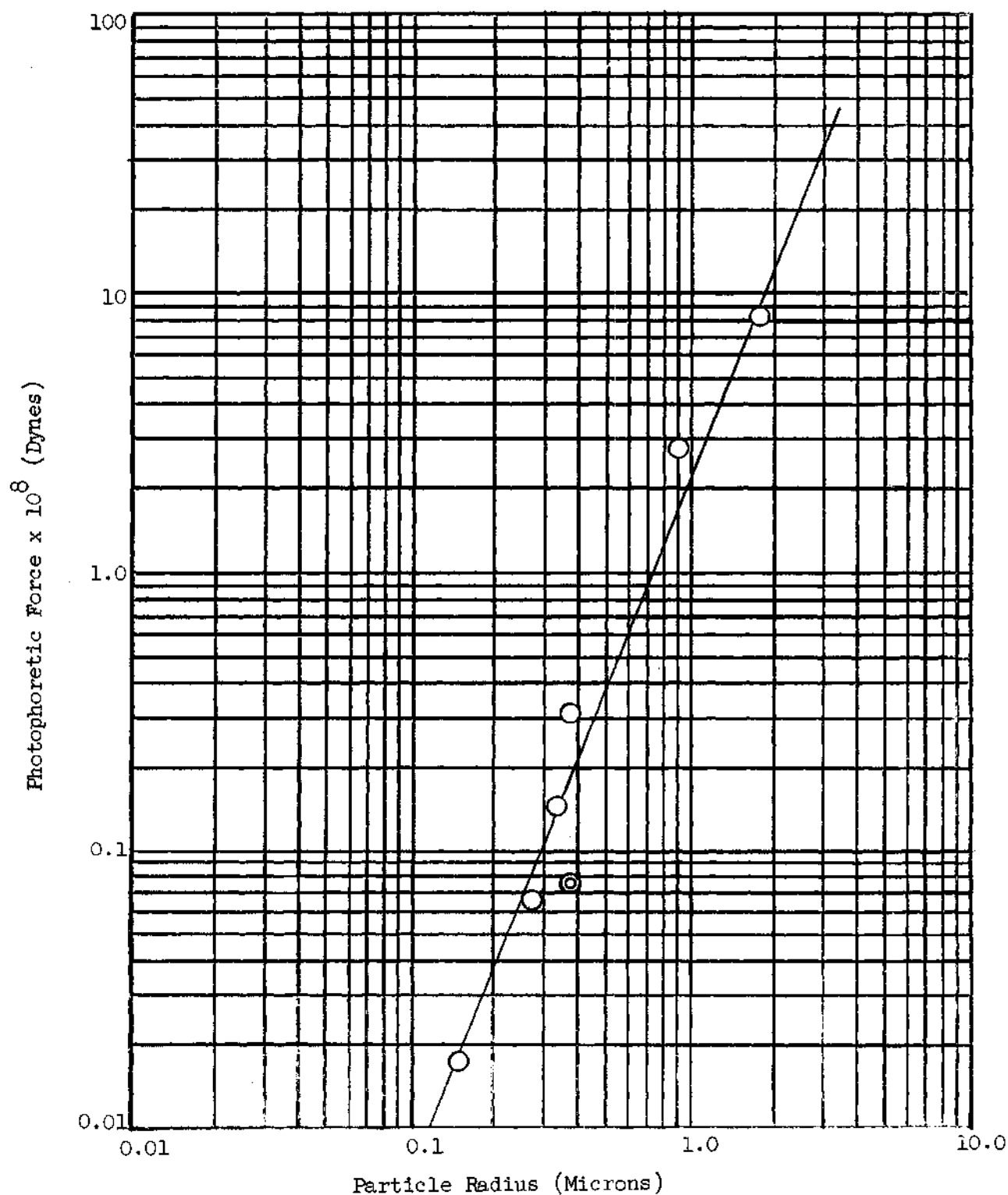


Figure 23. Negative Photophoretic Force versus Particle Radius for Sodium Chloride Particles at Pressures of 2 (⊙) and 10 (○) Torr.

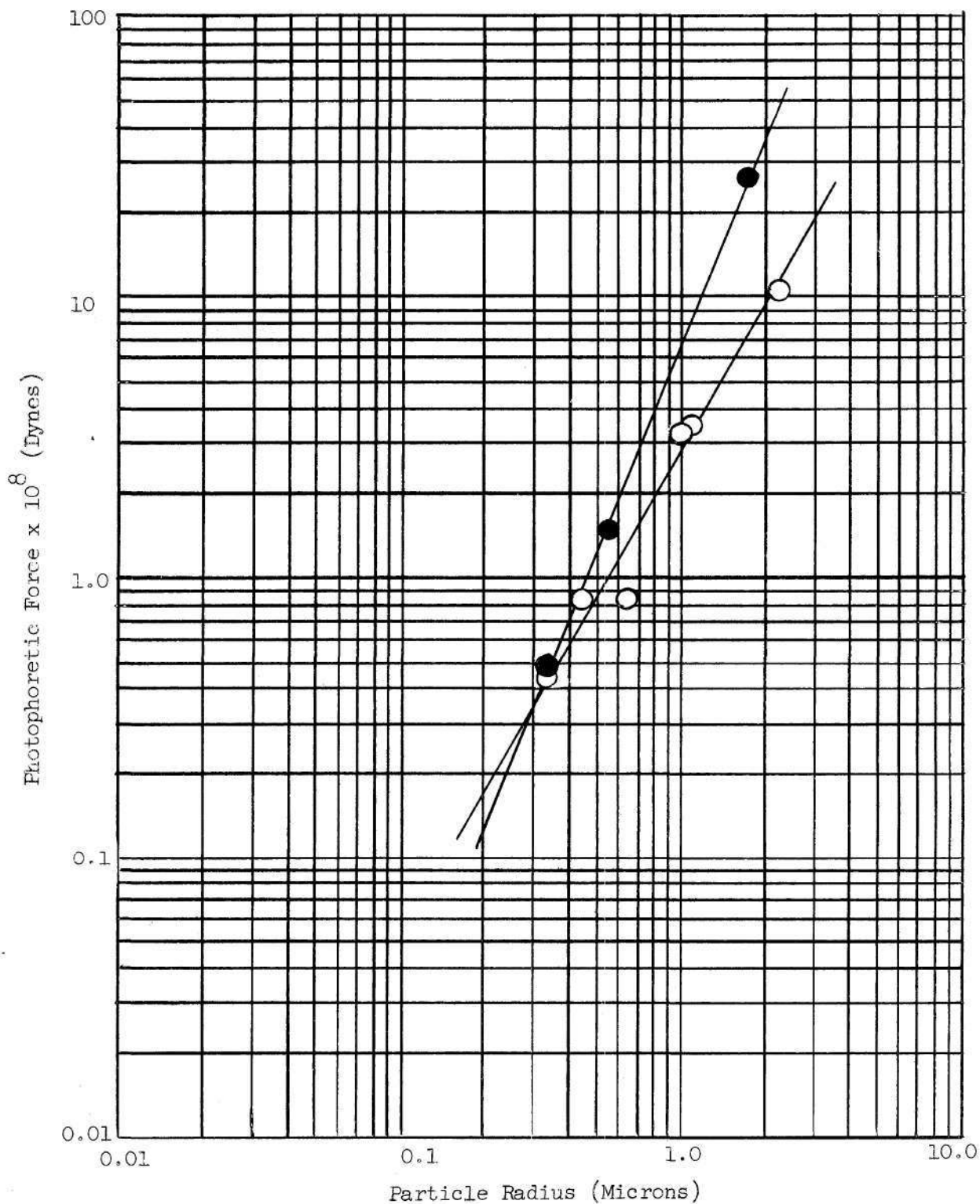


Figure 24. Positive (●) and Negative (○) Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 15 Torr.

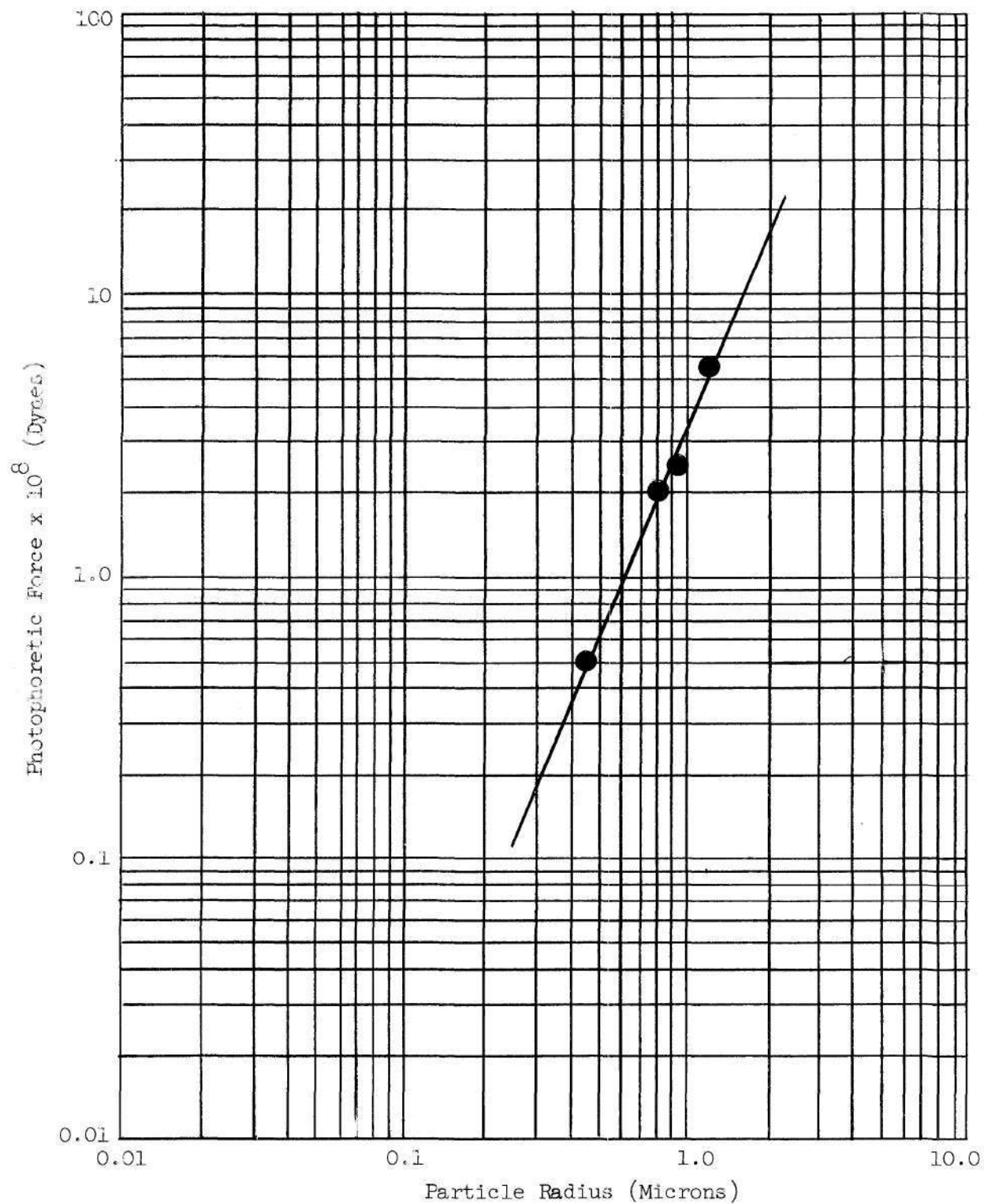


Figure 25. Positive Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 20 Torr.

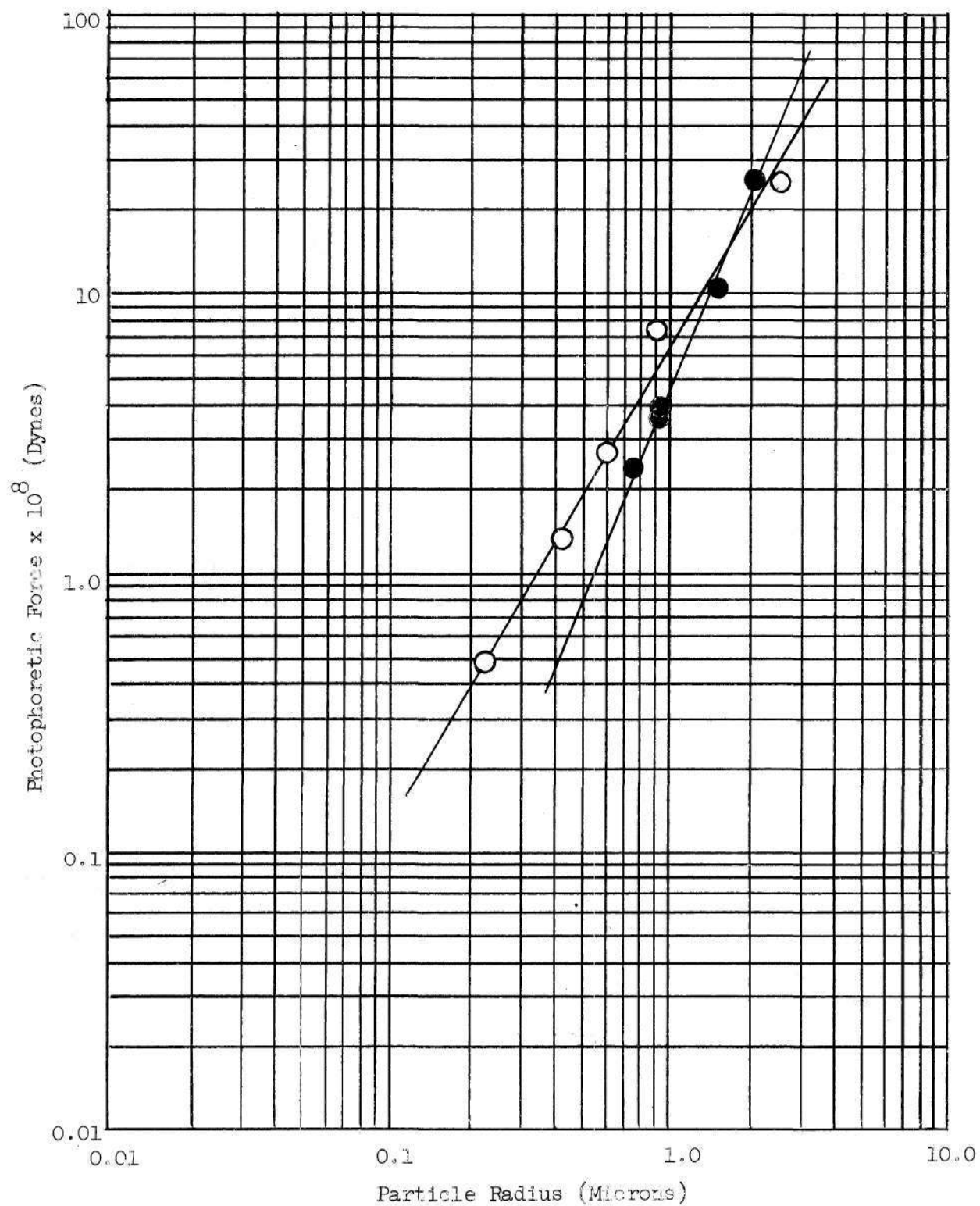


Figure 26. Positive (●) and Negative (○) Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 30 Torr.

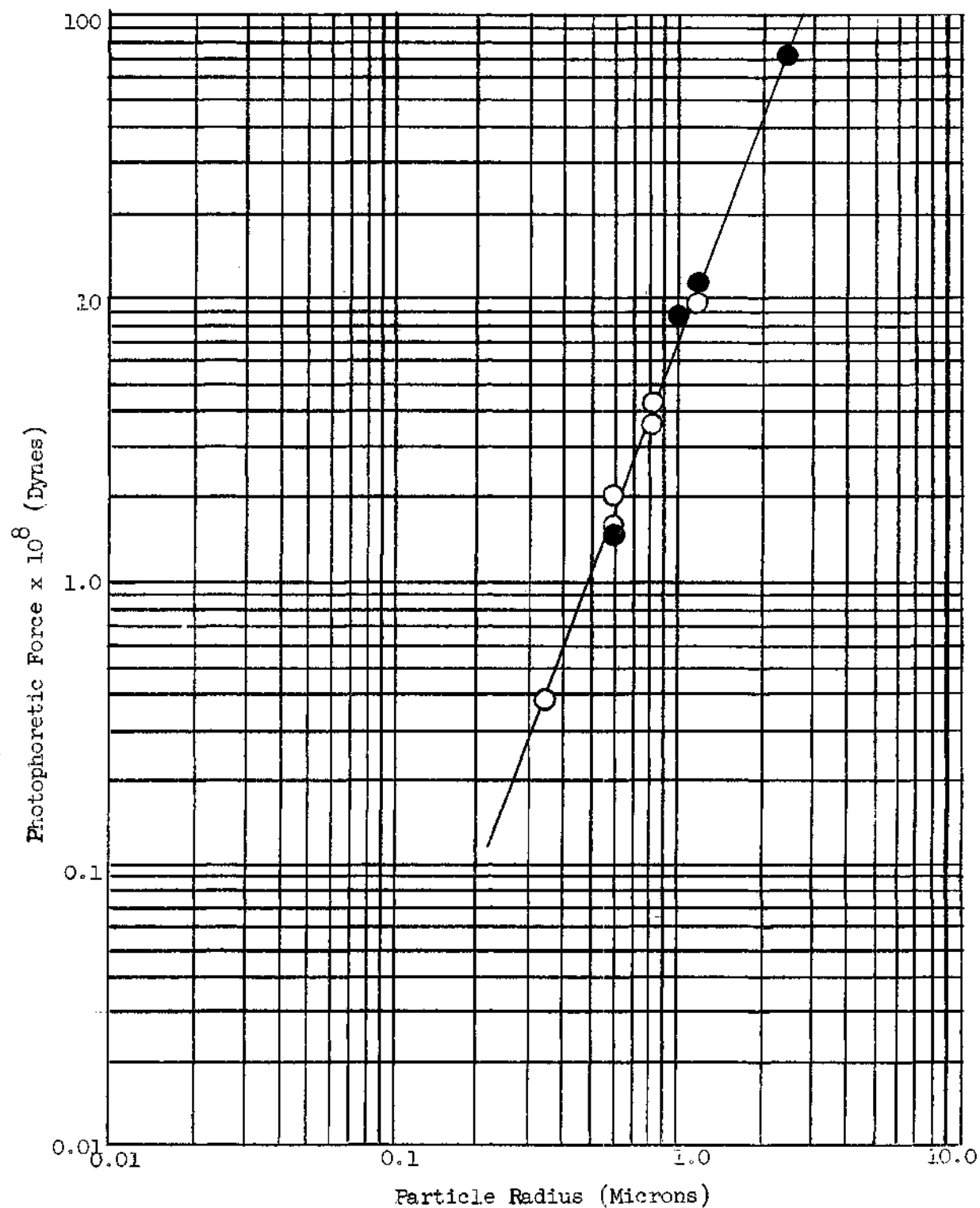


Figure 27. Positive (●) and Negative (○) Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 50 Torr.

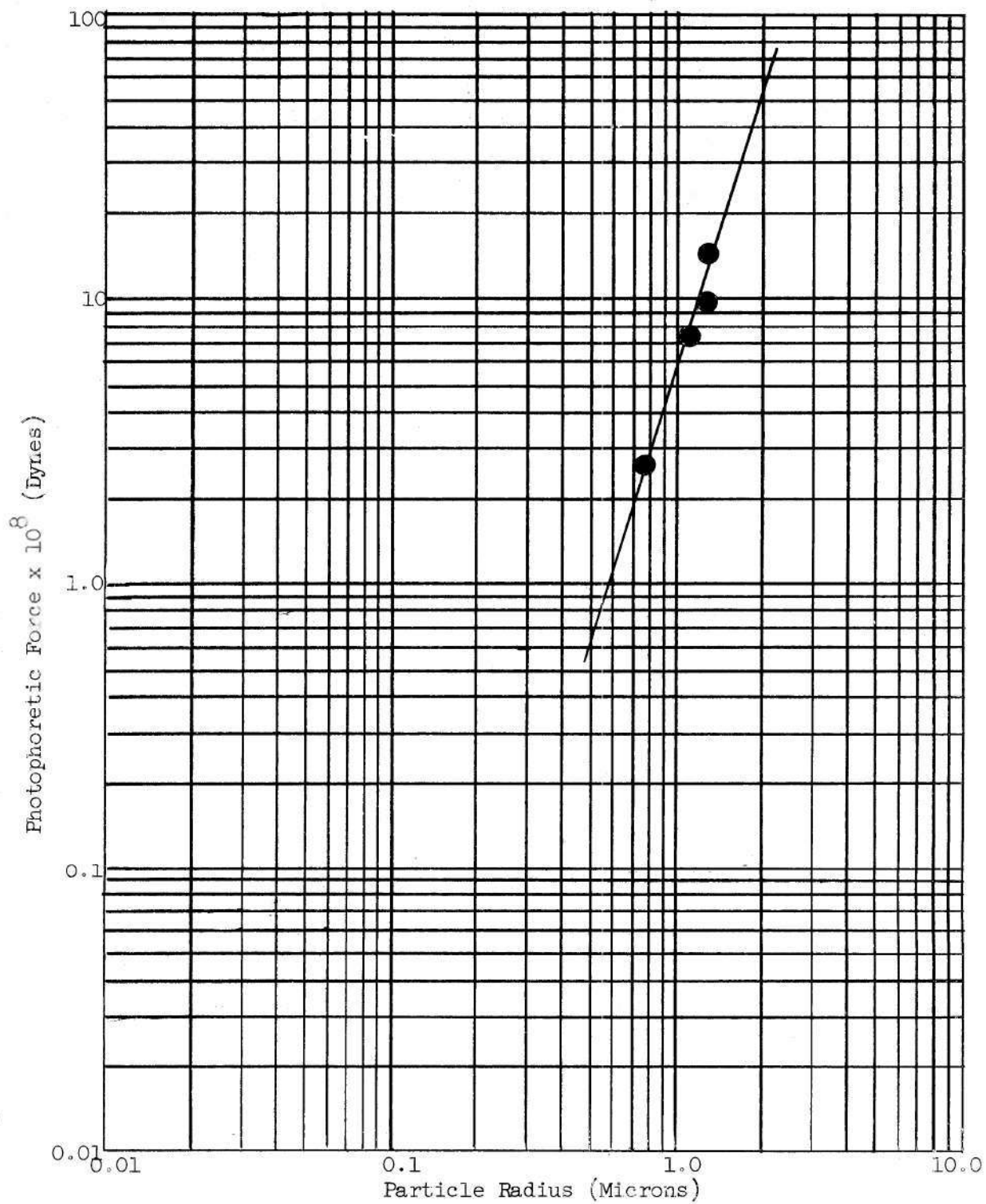


Figure 28. Positive Photophoretic Force versus Particle Radius for Sodium Chloride Particles at a Pressure of 63 Torr.

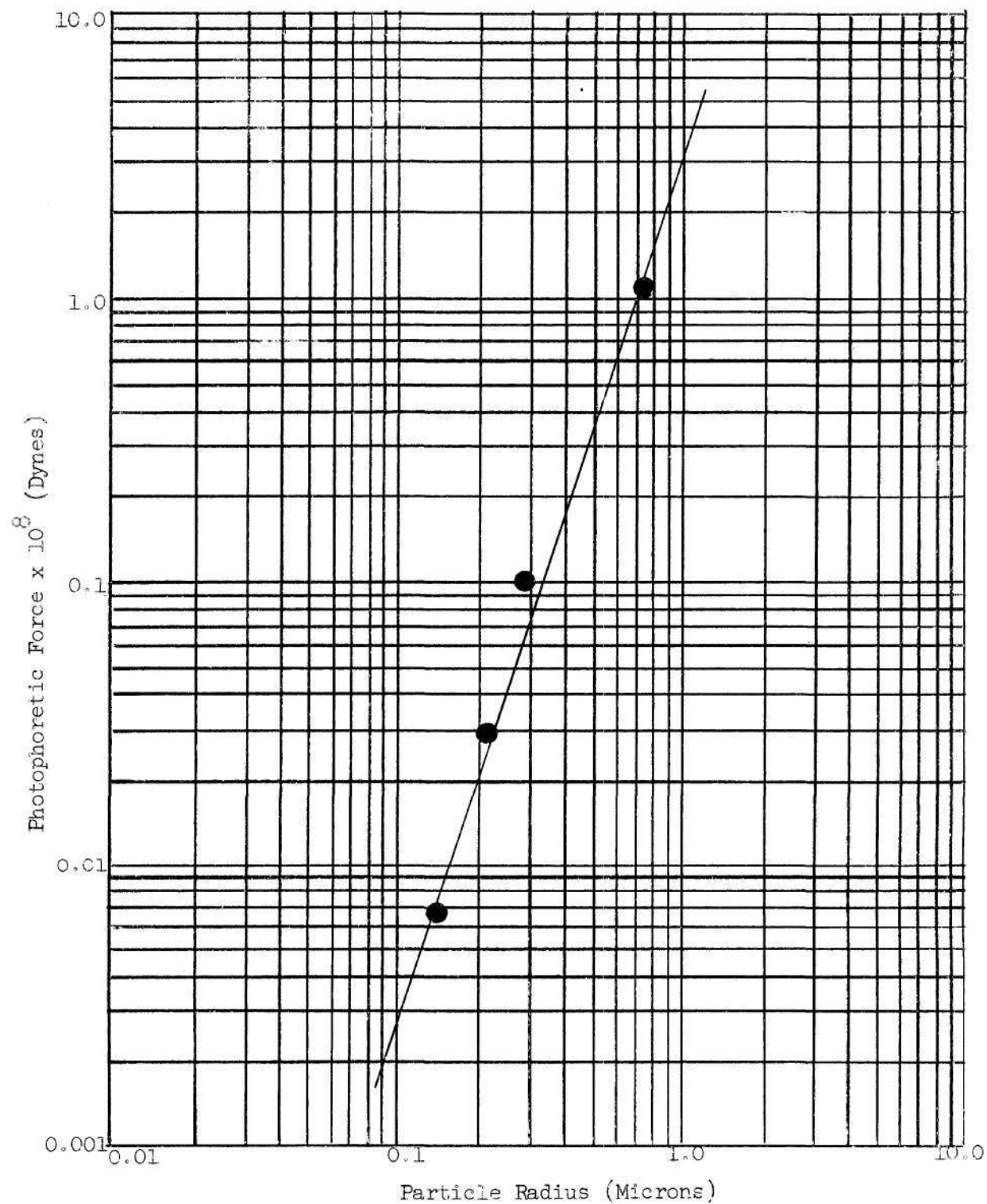


Figure 29. Positive Photophoretic Force versus Particle Radius for Silicon Carbide Particles at a Pressure of 2 Torr.



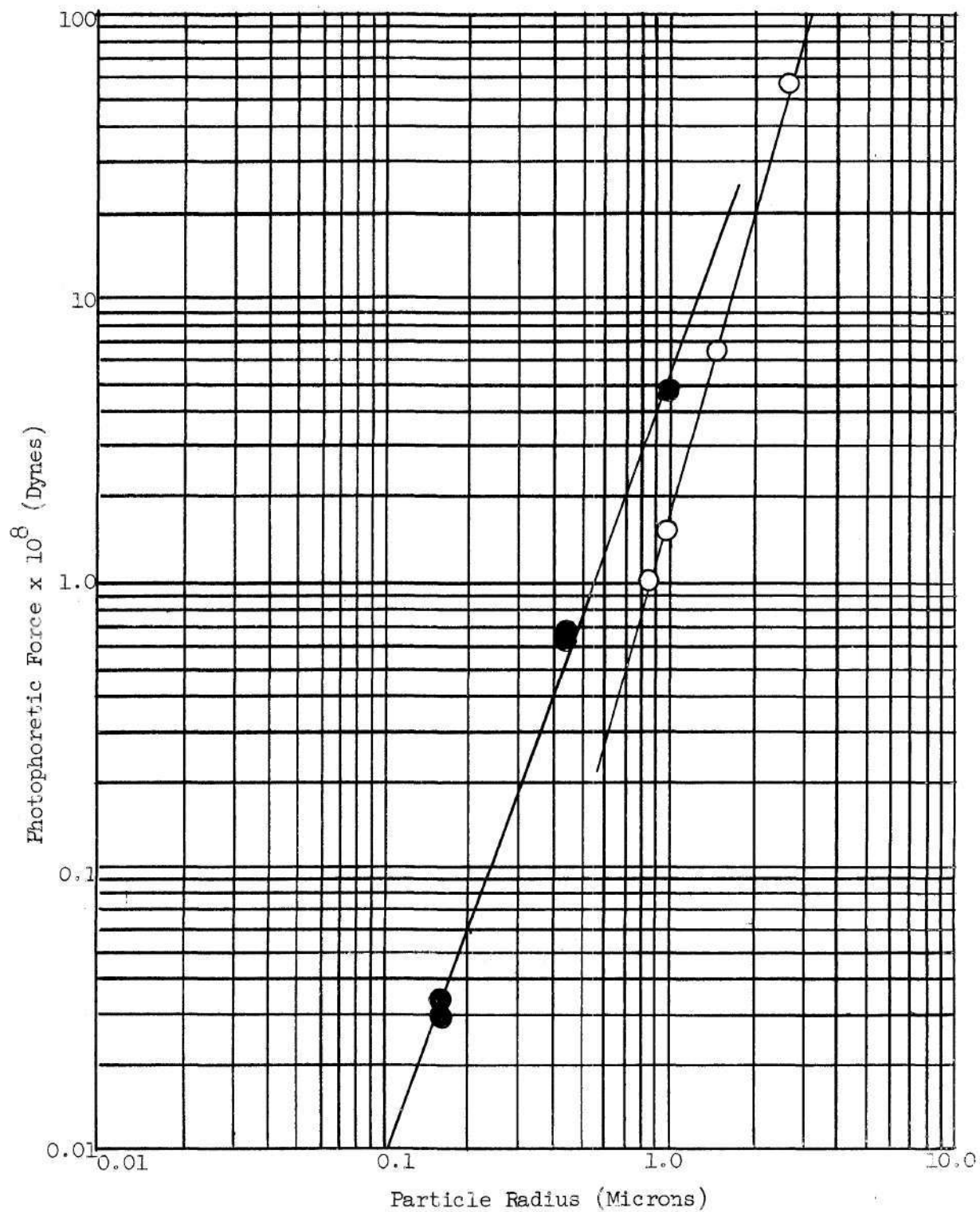


Figure 30. Positive (●) and Negative (○) Photophoretic Force versus Particle Radius for Silicon Carbide at a Pressure of 5 Torr.



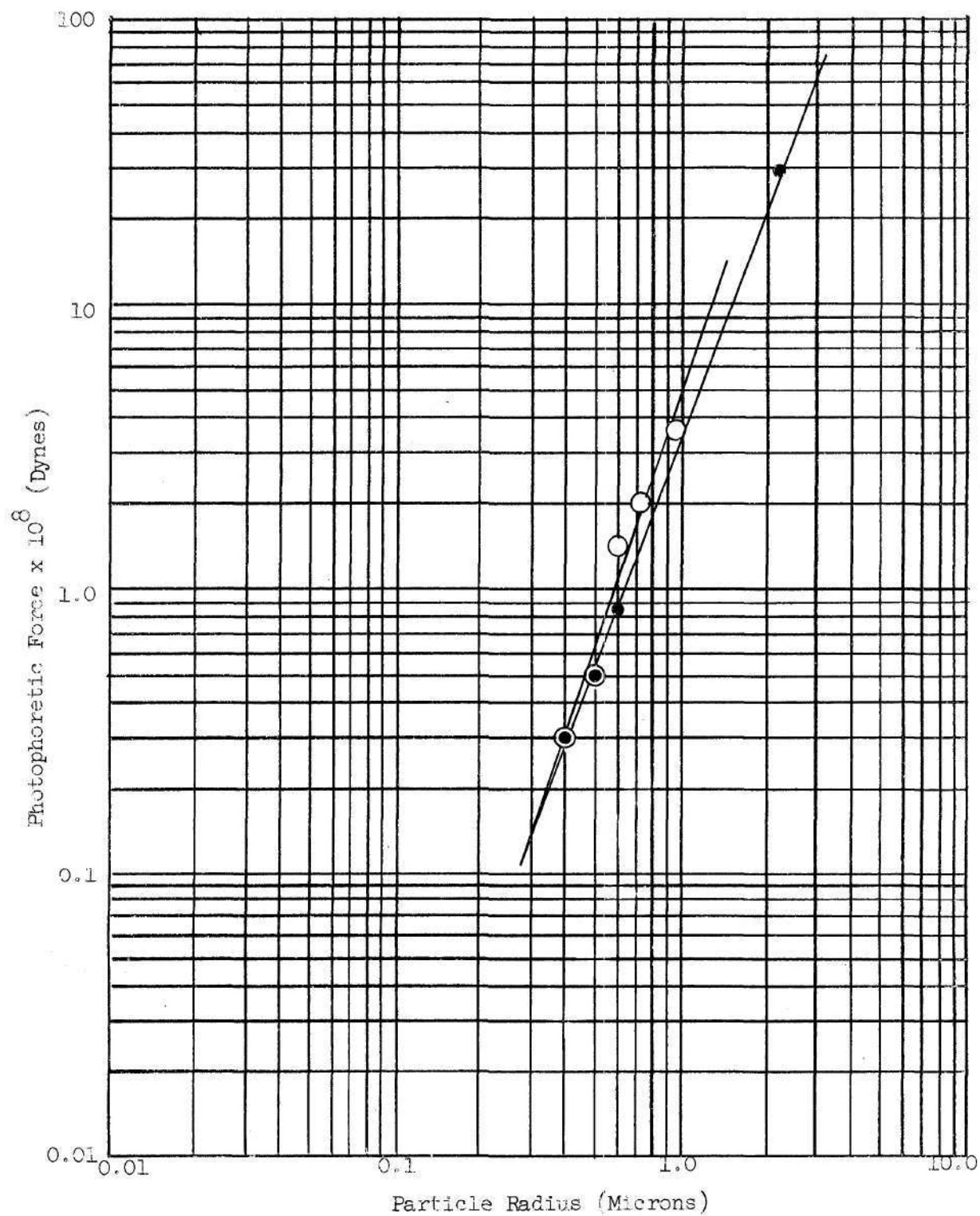


Figure 31. Positive (●) and Negative (○) Photophoretic Force versus Particle Radius for Silicon Carbide Particles at a Pressure of 10 Torr.

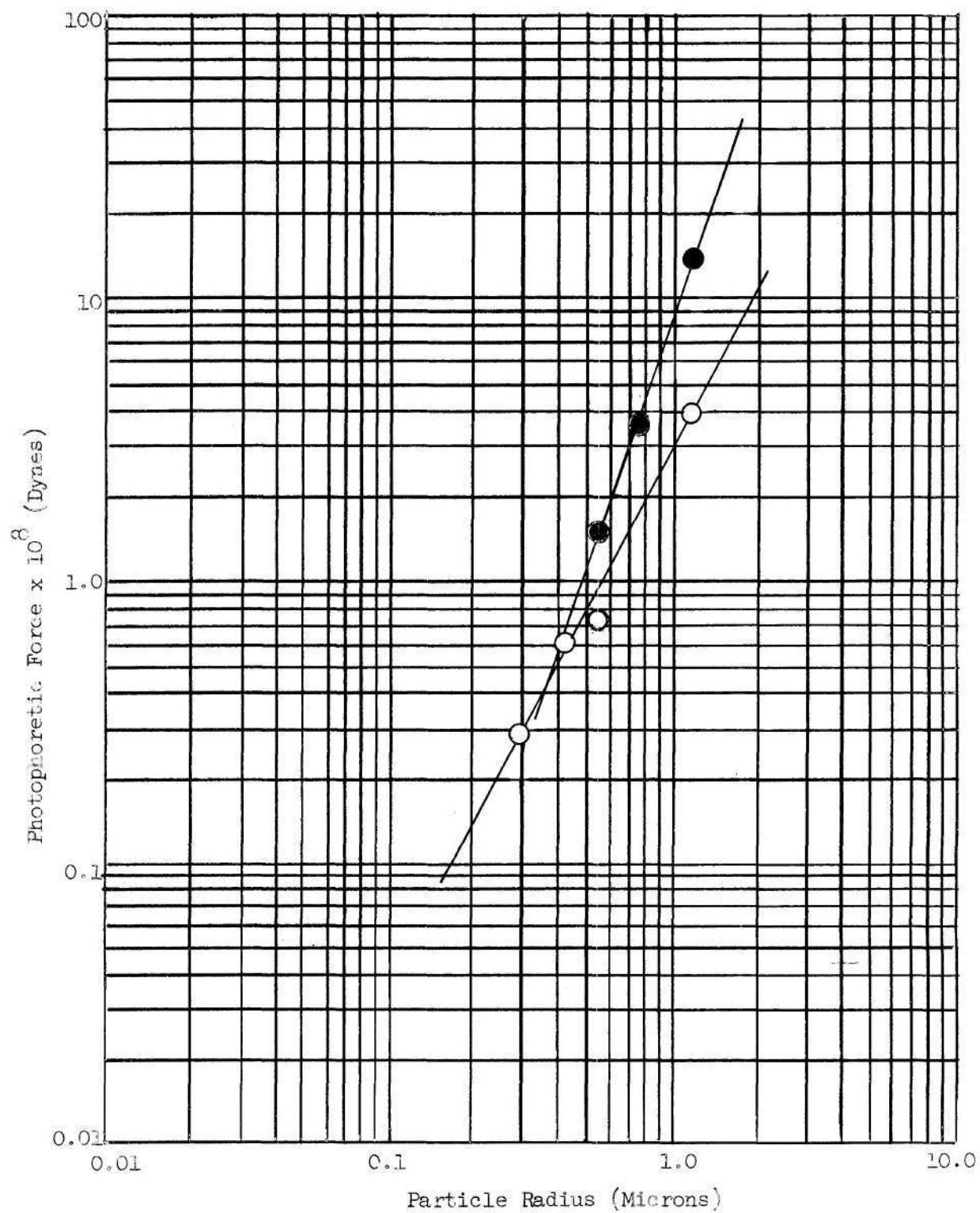


Figure 32. Positive (●) and Negative (○) Photophoretic Force versus Particle Radius for Silicon Carbide Particles at a Pressure of 30 Torr.

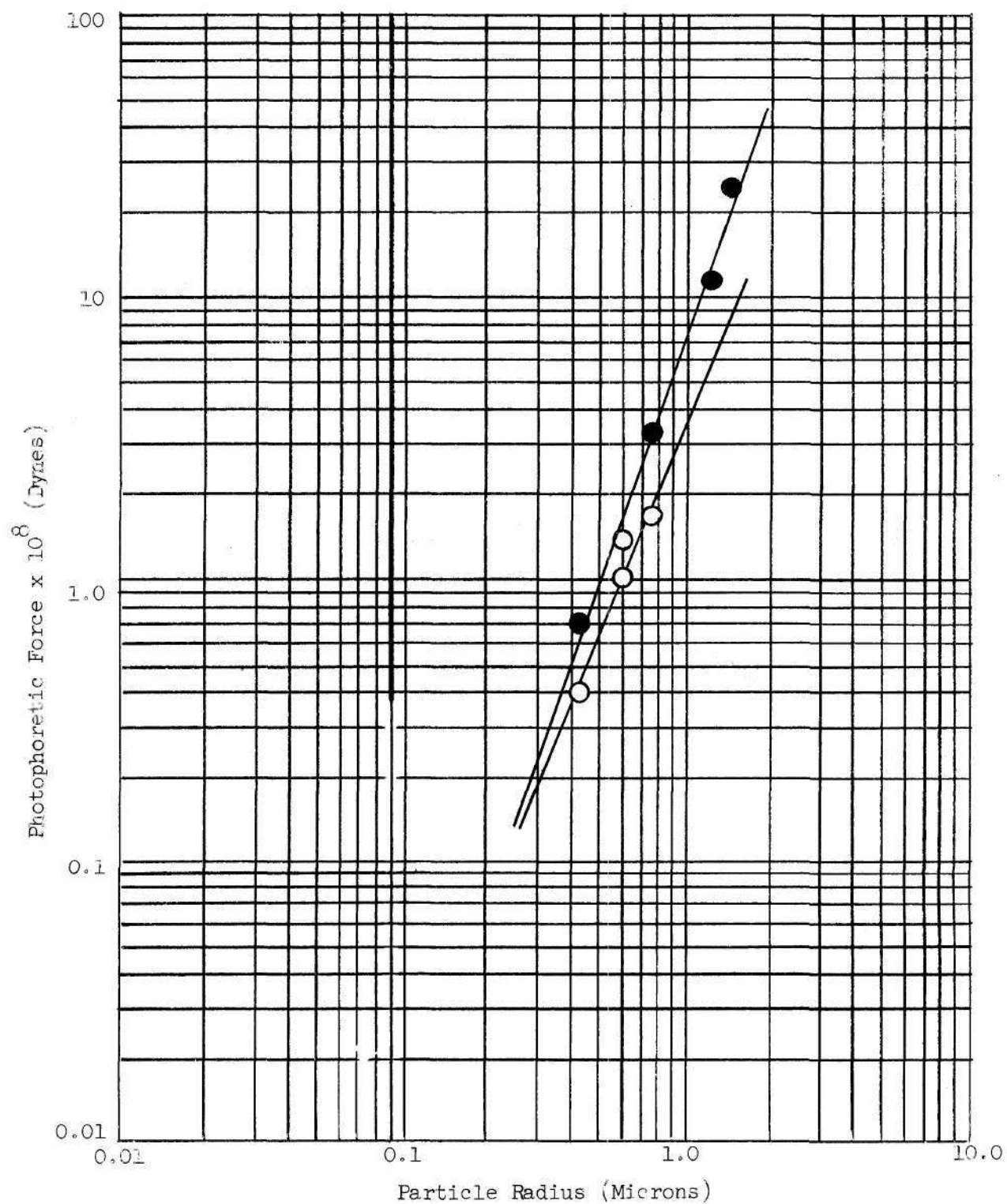


Figure 33. Positive (●) and Negative (○) Photophoretic Force versus Particle Radius for Silicon Carbide Particles at a Pressure of 50 Torr.

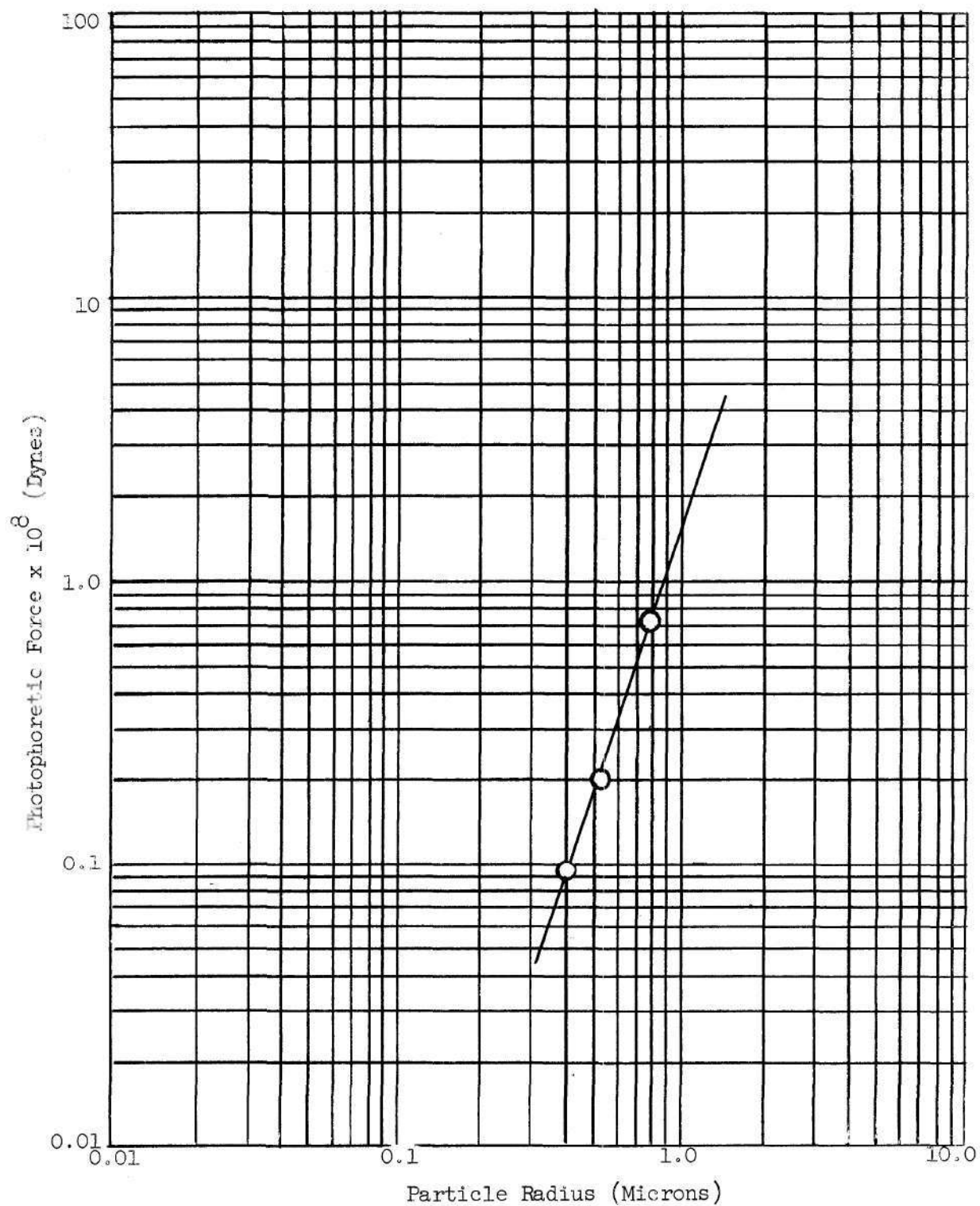


Figure 34. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 0.6 Torr.

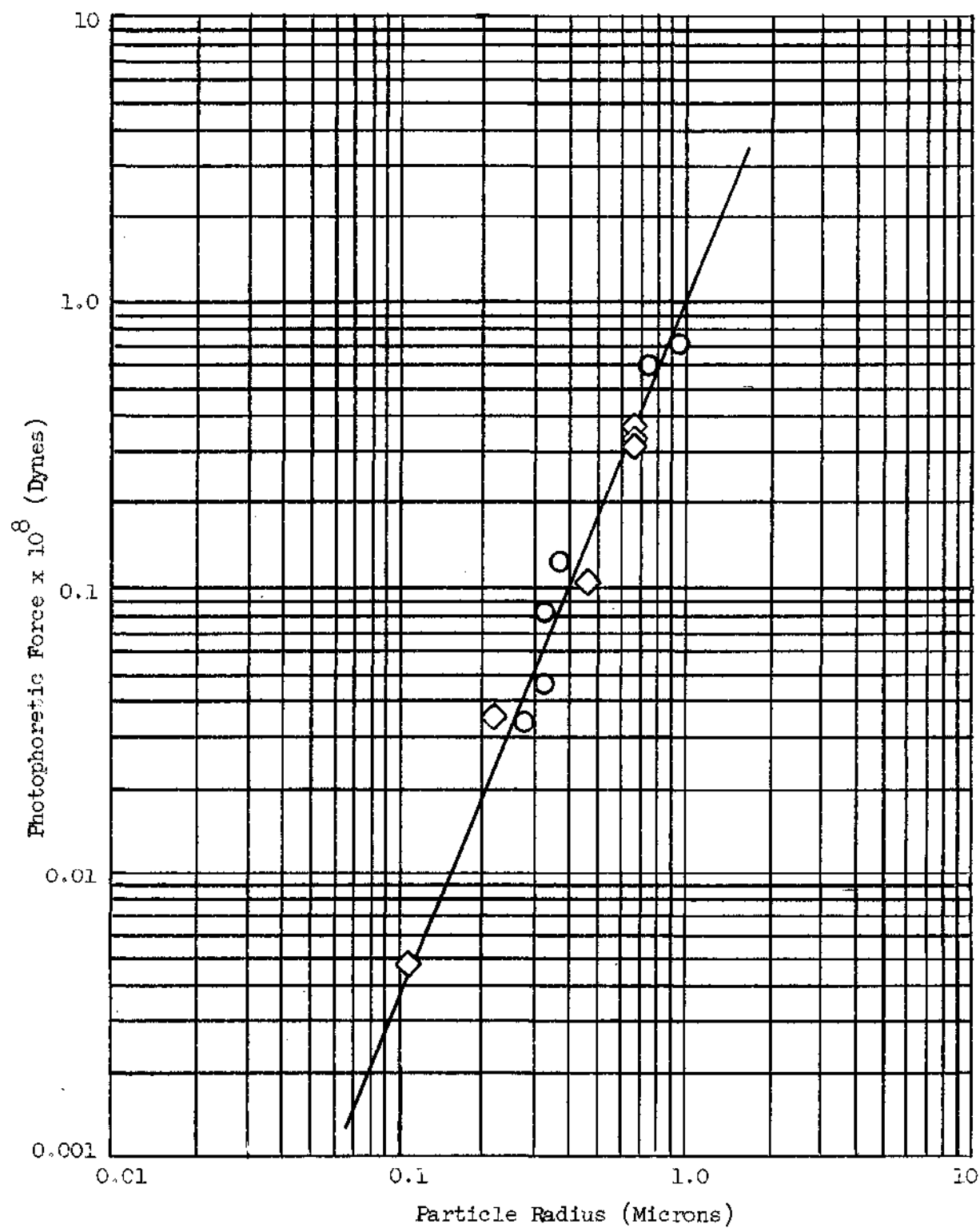


Figure 35. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 1.5 Torr in Air (○) and Helium (◇).

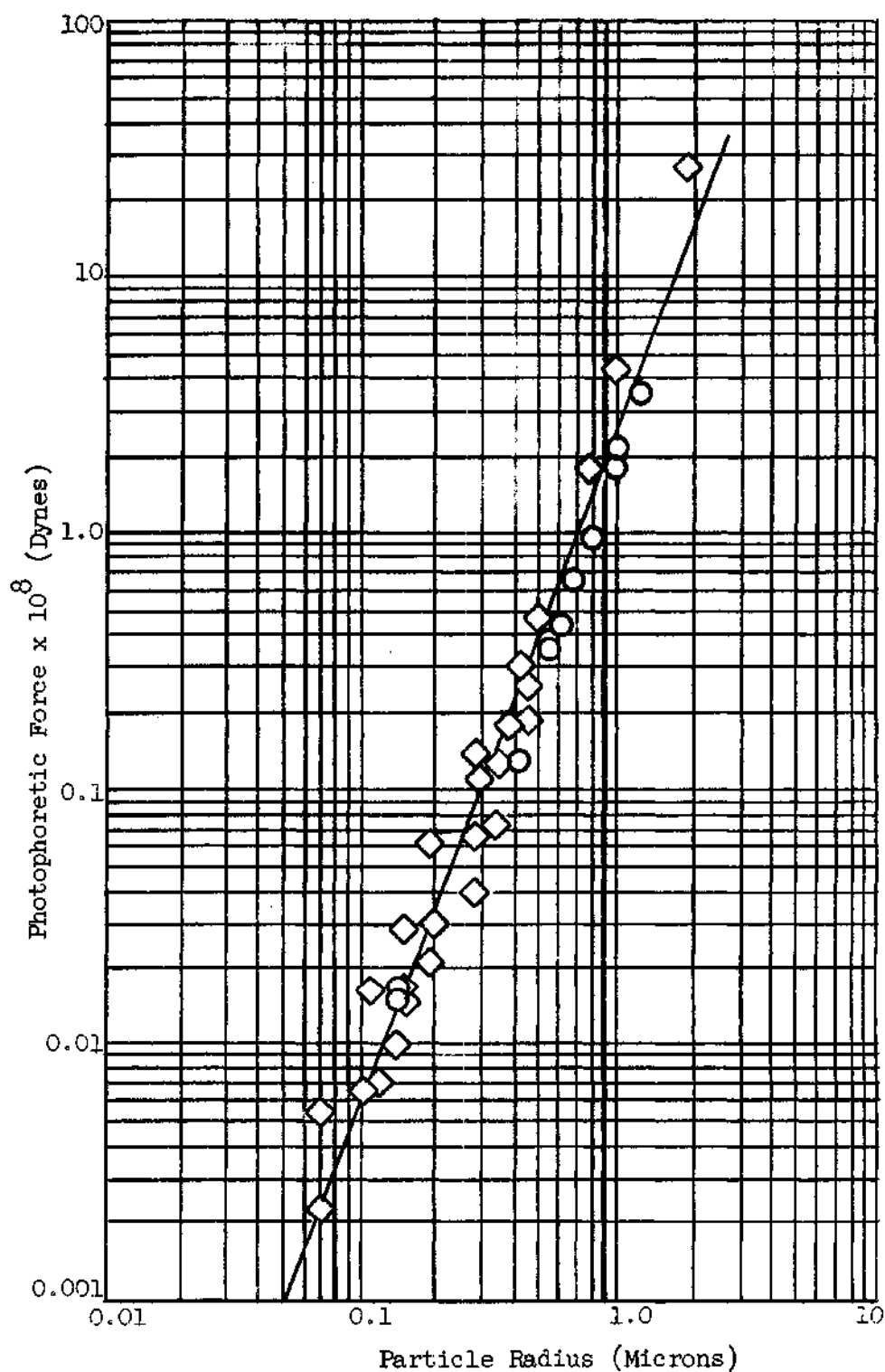


Figure 36. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 10 Torr in Air (○) and Helium (◇).

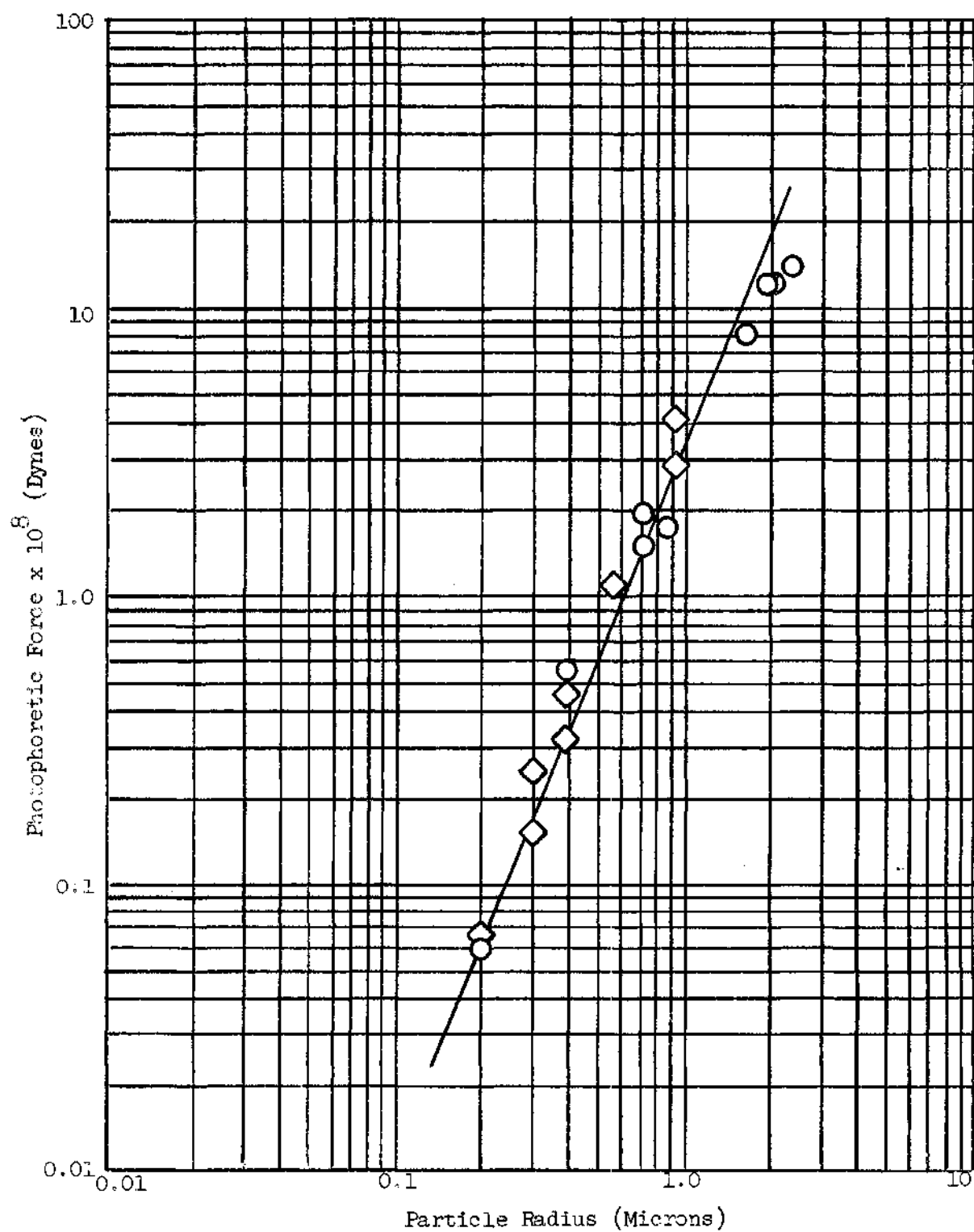


Figure 37. Negative Photophoretic Force versus Particle Radius for Carbon Particles at a Pressure of 30 Torr in Air (O) and Helium (◇).

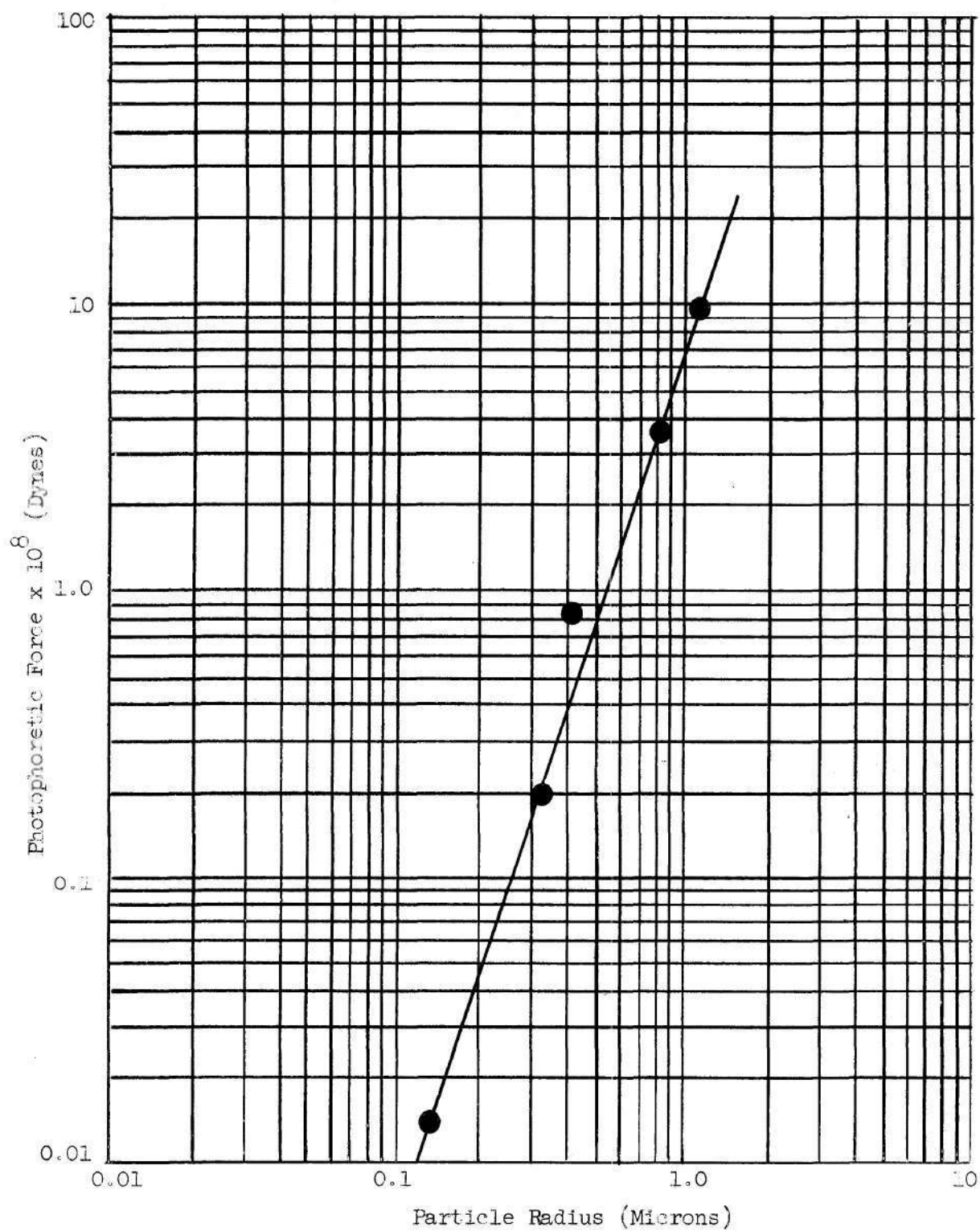


Figure 38. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 4 Torr.



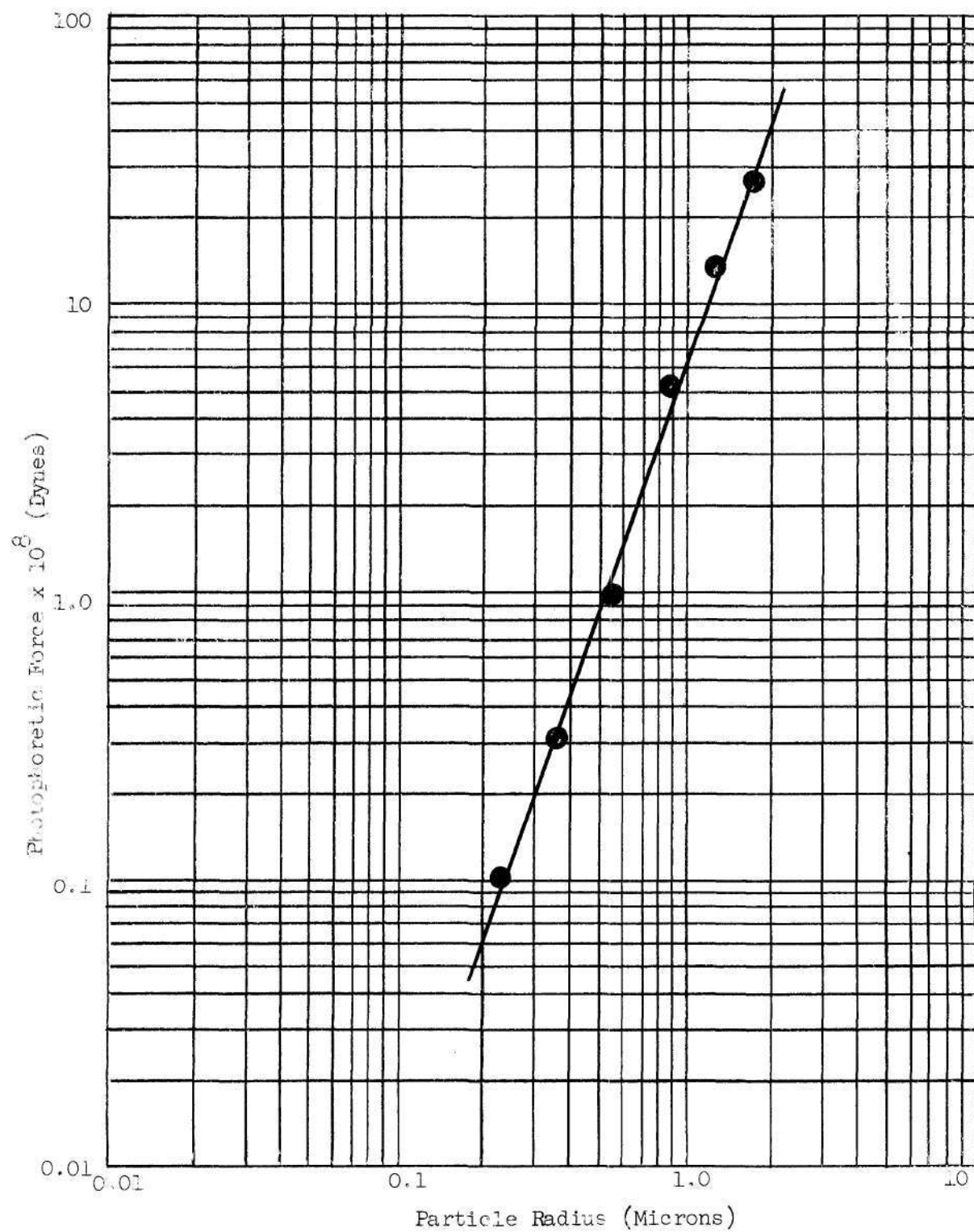


Figure 39. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 10 Torr.

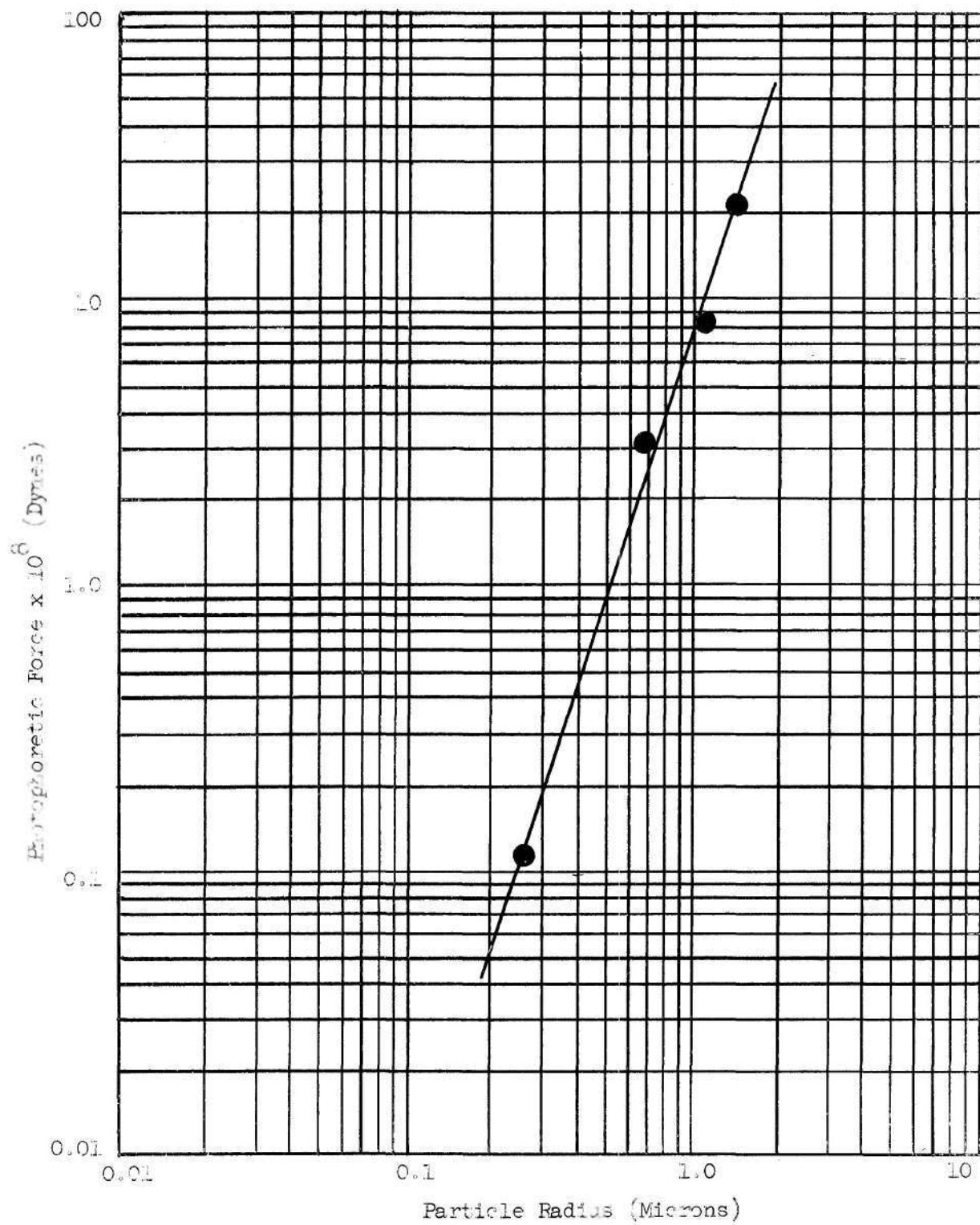


Figure 40. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 20 Torr.

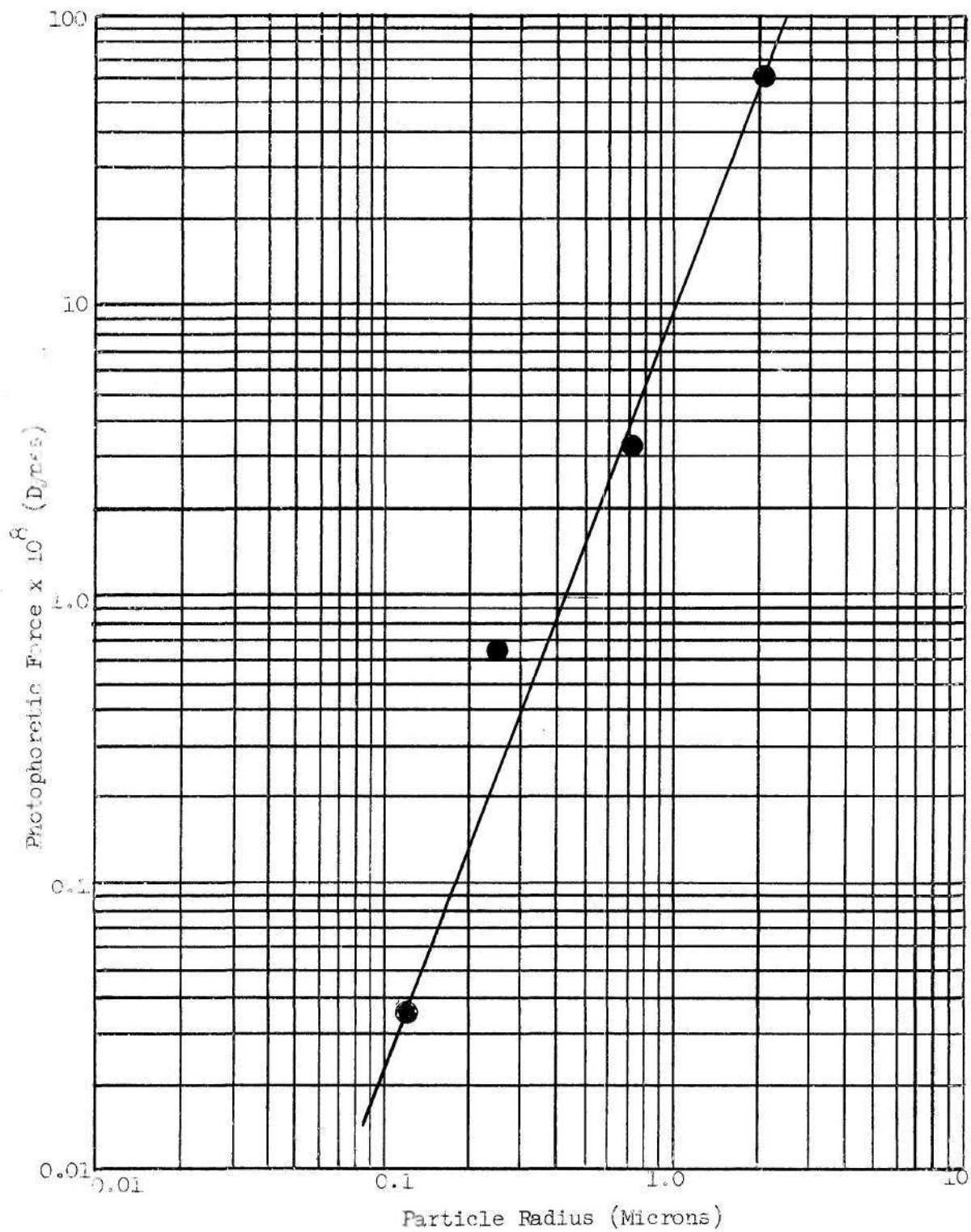


Figure 41. Positive Photophoretic Force versus Particle Radius for Zinc Particles at a Pressure of 30 Torr.

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